AD-A170 811

REPORT DOCUMENTATION PAGE

1a. REPORT SECURITY CLASSIFICATION Unclassified	1b. RESTRICTIVE MARKINGS
2a. SECURITY CLASSIFICATION AUTHORITY	3. DISTRIBUTION/AVAILABILITY OF REPORT Approved for public release, distribution unlimited
2b. DECLASSIFICATION/DOWNGRADING SCHEDULE	
4. PERFORMING ORGANIZATION REPORT NUMBER(S) Yale University New Haven, Connecticut 06520	5. MONITORING ORGANIZATION REPORT NUMBER(S) AFOSR·TR· 86-0522

6a. NAME OF PERFORMING ORGANIZATION Yale University	6b. OFFICE SYMBOL (If applicable)	7a. NAME OF MONITORING ORGANIZATION AFOSR/NP
6c. ADDRESS (City, State and ZIP Code) New Haven, CT 06520		7b. ADDRESS (City, State and ZIP Code) Building 410 Bolling HFB, DC 20332-6448
8a. NAME OF FUNDING/SPONSORING ORGANIZATION AFOSR	8b. OFFICE SYMBOL (If applicable) NP	9. PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER AFOSR-81-0077

8c. ADDRESS (City, State and ZIP Code) Bolling Air Force Base Washington, D.C. 20332	10. SOURCE OF FUNDING NOS.			
	PROGRAM ELEMENT NO. 61102F	PROJECT NO. 2301	TASK NO. A8	WORK UNIT NO.

11. TITLE (Include Security Classification)
Population Inversions in Laser-Initiated Vacuum Arcs

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Gary L. Haller** (Chairman, Council of Engineering) P.I. 9/1/85-1/31/86

13a. TYPE OF REPORT Final	13b. TIME COVERED FROM 2/1/81 TO 1/31/86	14. DATE OF REPORT (Yr., Mo., Day) 17 June 1986	15. PAGE COUNT

16. SUPPLEMENTARY NOTATION

AUG 13 1986

17. COSATI CODES			18. SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB. GR.	

19. ABSTRACT (Continue on reverse if necessary and identify by block number)

Population inversions were studied in laser-initiated vacuum arcs. Inversion mechanisms include three-body recombination pumping in expanding, laser produced plasmas, and resonant photoexcitation using line radiation. Significant gain was measured on the Balmer lines H_α and H_β in expanding, laser produced plasmas of carbon/polyethylene. Research then focused on resonant photoexcitation of one ion by line radiation from another ion. Fluorescence was measured at UV wavelengths in CII pumped by Al III. A new class of photoexcited lasers was identified, in Be-like ions, with wavelengths from 2177Å in C III pumped by Mn VI, to 200Å in Mg IX pumped by Al XI. Fluorescence and small-signal gain were measured in C III pumped by Mn VI. Finally, laser oscillation was demonstrated in C III at 2177 and 2163Å. These are the first UV lasers to be pumped by resonant photoexcitation. By isoelectronic scaling, it should be possible to produce soft X-ray lasers using the principles elucidated by this research. Photopumped lasers could prove to be the most efficient soft X-ray lasers in the future.

20. DISTRIBUTION/AVAILABILITY OF ABSTRACT UNCLASSIFIED/UNLIMITED ☑ SAME AS RPT. ☐ DTIC USERS ☐	21. ABSTRACT SECURITY CLASSIFICATION
22a. NAME OF RESPONSIBLE INDIVIDUAL	22b. TELEPHONE NUMBER (Include Area Code) 415:577-7126	22c. OFFICE SYMBOL NP

DD FORM 1473, 83 APR EDITION OF 1 JAN 73 IS OBSOLETE. UNCLASSIFIED

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17 June 1986

Dr. Robert J. Barker
Office of Scientific Research
United States Air Force
Building 410
Bolling AFB, D.C.   20332

Dear Dr. Barker:

    Please find enclosed the Final Report on AFOSR Contract 81-0077, prepared by Dr. Mahadevan Krishnan.   I believe this report completes Dr. Krishnan's obligation to the contract.

Sincerely yours,

Gary L. Haller
Becton Professor
and Chairman

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I. __INTRODUCTION__

This is the final Technical Report for AFOSR Grant #81-0077, covering the 60 month period from Feb.1,1981 to Jan.31,1986. The total dollar amount awarded was $479,191. No cost overruns were incurred during the entire period. Eight journal publications have already resulted from the research supported by this Grant, including two Phys. Rev. Letters and two App. Phys. Letters. Two more publications are in preparation. Two Ph.D theses were completed under this Grant. One postdoctoral student and several graduate and undergraduate students were trained in the course of this research. The technical scope of this Grant evolved towards the study of resonant photo-excitation in arc discharges, pumped by line radiation from laser-produced plasmas. The Ph.D thesis of James Trebes studied the basic physics of CII pumped by AlIII. These studies identified a new class of photo-excited lasers in Be-like ions, from CIII to MgIX. The Ph.D thesis of Niansheng Qi focused on CIII pumped by MnVI. Small signal gain was measured on several transitions in CIII, from 1620 to 2177Å. Finally, laser oscillation was demonstrated at 2163 and 2177Å. These are the first UV lasers to be pumped by resonant photo-excitation. By isoelectronic scaling, it is possible to pump soft X-Ray lasers using the same principles. High overall efficiency lasers could result. The rest of this report provides more detail on the progress of this research.

II. RESEARCH OBJECTIVES

The overall objective of this research program was to study population inversions in laser-initiated vacuum arcs. Early in the program, the emphasis was on three-body recombination in expanding, laser-produced plasmas of carbon and carbon/polyethylene. It had already been established by others that rapid cooling due to expansion leads to three-body recombination, which populates the high lying states of the recombined ions. Subsequent collisional-radiative cascades then produce population inversions. The gains measured in laser-produced plasmas were too low, because the rapid cooling of these plasmas was accompanied by a rapid decrease in density, which accounted for the low gain. The objective of the proposed program at Yale was to inhibit this decrease in density by confining the expansion. Ideally, it is desirable to allow rapid cooling of the plasma, but without decreasing the density significantly. To achieve this, it was proposed to run a high current discharge between the laser target and an anode electrode. The self-consistent electric and magnetic fields of the discharge would confine the expansion of the laser-produced plasma, thereby allowing for higher gain due to recombination pumping.

After the first two years, the focus of the research changed to the study of resonant photo-excitation. The revised objective now was to study the detailed physics of photo-excitation in one ion by line radiation from another ion. The studies conducted at Yale at UV wavelengths were to provide scaling laws for soft X-Ray lasers in higher Z, isoelectronic analogs. As stated in the introduction, significant progress was made in this regard. The next section highlights the progress.

III. PROGRESS

Many aspects of the research performed under this Grant are well summarized in journal publications. However, early work on population inversions due to three-body recombination have not been published in the archival literature. This section begins by reviewing this research. The subsequent photo-excitation work is reviewed by including selected papers as appendices.

III-1: Three-body recombination.

Prior to the start of this research, optical gain had been observed on the Balmer-α and Balmer-β lines of atomic hydrogen, in expanding, laser-produced plasmas from carbon/polyethylene targets. To characterize this gain phenomenon, experimental and theoretical work was carried out. The Stark broadening of the H_β line was used to measure the electron density as a function of distance from the laser target, and as a function of time, at a fixed distance. Typical results are shown in Fig.1, where the data are the full circles. The three curves on the figure are from a self-similar, hydrodynamic expansion model, for different assumed initial temperatures of 0.5 to 1.4eV. Because these temperatures are lower than expected for a 15J/100ns laser-produced plasma, it was concluded that the atomic hydrogen probably originated from a secondary plasma produced by the long(1.0μs) tail of the CO_2 laser. This secondary plasma would begin at a lower temperature and expand into vacuum more nearly spherically. Using these densities and electron temperatures, a multi-level, collisional-radiative model was developed for the

kinetics in the recombining hydrogen atoms. Typical results are shown in Fig.2. The figure plots the level populations of the $n=2$ to $n=6$ levels of hydrogen, for an initial electron density $n_0=10^{19}$ cm^{-3} and initial temperature $T=10$eV. The hydrodynamics of the expanding plasma were solved by dividing the initial sphere of radius R_0 into a number of concentric shells. The temporal evolution of each shell is then described by conservatic. equations, with an assumed ideal gas equation of state. The position, velocity, density and temperature of a shell initially centered at radius r_0 within the sphere are calculated as functions of time. Following the temporal evolution of many such shells then permits an Eulerian view of the flow. At each time step, these computed densities and temperatures were used as rate controlling inputs to a collisional-radiative model. For each quantum level, collisional excitation and de-excitation, three-body and radiative recombination, spontaneous decay and ionisation were included. The changing density and temperature led to time-dependent poulations in the levels, as shown in Fig.2. The level populations are shown as functions of time over the first $0.5\mu s$ interval following the laser pulse. Also shown on the abscissa are the spatial locations at each time, of the particular shell of plasma which is being followed. For these conditions, the population of $n=1$ greatly exceeds that of the higher levels i.e. no inversion is predicted for the Lyman lines. However, the figure does show that over almost the entire duration, the four Balmer lines $H_\alpha, H_\beta, H_\gamma$ and H_δ are all inverted. Two features of the collisional-radiative code developed here are worth noting: first, many earlier attempts[1] assumed a quasi-steady state for the upper level populations. As Fig.2 shows, this is clearly a dynamic process in which all levels are time-varying. Second, plasma dynamics are included in this model by retaining the convective derivative, $V.V$, in the individual rate equations. The

1. U.Furukane, T.Yokoto, and T.Oda, JQSRT, 22, 239 (1979)

convective velocity field in turn is obtained from the hydrodynamic solution.

Fig.3 shows the measured small-signal gain (using single-pass reflection) at H_α. The first 15mm from the target show no gain, although the theory predicts gain. This discrepancy was accounted for by considering the opacity of the Lyman lines, which effectively destroys the inversion on the Balmer lines.

The research into three-body recombination pumped inversions was quickly superceded by the photo-excitation research. The rather high gains measured in hydrogen Balmer lines suggest that a hydrogen laser might be developed, if the work was pursued further. Such an atomic hydrogen laser would have been the first of its kind, and might offer some applications for neutral diagnostics in fusion plasmas.

III-2:   Resonant Photo-excitation

As stated in the introduction, resonant photo-excitation research at Yale began with the study of the CII-AlIII scheme. A detailed study of this scheme resulted in the Ph.D thesis of James Trebes. A comprehensive summary of this work is contained in a paper- "UV Fluorescence by Optical Pumping with Line Radiation", by James Trebes and Mahadevan Krishnan, which was published in Laser Techniques in the Extreme Ultraviolet (OSA, Boulder, Colorado,1984), S.E.Harris and T.B.Lucatorto, Eds. (AIP, New York, 1984), p.387. Because this final report attempts to give a comprehensive account of the entire 5-year research program, the above paper is included, as Appendix I. One outgrowth of Trebes' thesis was the identification of a new class of photo-excited lasers in Be-like ions, from CIII to MgIX. These lasers offer wavelengths from 2177Å in CIII to ~200Å in MgIX. The

proposed lasers were described in -"Proposed New Class of Optically Pumped, Quasi-cw, Ultraviolet and Extreme Ultraviolet Lasers in the Be Isoelectronic Sequence", M.Krishnan and J.Trebes, Appl. Phys. Lett.45, 189 (1984). By June of 1984, we had already observed significant fluorescence in CIII pumped by MnVI line radiation. These measurements were followed by measurements of small signal gain at 2177 and 2163Å in CIII. The gain measurements were compared with the predictions of a 72-level, collisional radiative code for CIII. In developing this code, we were fortunate to have the support of Dr.W.L.Morgan of Lawrence Livermore Labs., who graciously provided us with a complete set of energy levels, oscillator strengths, collisional and radiative rates for CII, CIII and CIV. Electron density and temperature were measured as functions of distance along the carbon discharge axis. These were then the rate controlling inputs to the atomic physics code, which calculated the various level populations. Optical pumping was modelled by assigning a brightness temperature of 20eV to the Mn pump line. The enhanced populations of the n=4 and n=3 levels, and hence the expected small-signal gain, were then calculated. Good agreement was obtained between code and experiment. Finally, the discharge geometry was modified to maximize the optical path length for gain. This led to the clear demonstration of lasing at 2177 and 2163Å. This detailed succession of experiments and theory forms the basis of the Ph.D thesis of Niansheng Qi. Three papers provide a summary of these developments:

Observed Enhanced Fluorescence at 2177,2163,1923 and 1620Å in CIII by Photo-excitation with MnVI Line Radiation at 310Å," N.Qi, H.Kilic, and M.Krishnan, Appl.Phys.Lett. March,1985

"Fluorescence and Small-signal Gain at 2177 and 2163Å in CIII by Resonant Photo-excitation with 310Å, MnVI Line

Radiation," N.Qi, H.Kilic, and M.Krishnan, Journal de Physique, to be published (Aug.1986)

"Laser Oscillation at 2177 and 2163Å in CIII, Resonantly Photo-excited by 310Å, MnVI Line Radiation," N.Qi and M.Krishnan, Phys.Rev. Lett. (accepted for publication)

For completeness, these papers are included in this report as Appendices II, III, and IV.

The demonstration of lasing in CIII bodes well for the success of such photo-excitation schemes in higher Z, isoelectronic analogs. A natural extension of the Yale work would be to show lasing in NIV pumped by PIX, with wavelengths of 900-1284Å. Also, such schemes might be tried using pulse-power driven Z-pinches, which can be efficient sources of pump radiation. To date, no support for pursuing these schemes has emerged from federal agencies. Prof.H.Griem of the University of Maryland has offered to pursue this line of research in his laboratory. Yale University is amenable to the transfer of the major items of equipment to Maryland, to continue the research. If funding is available, these avenues will be explored.

IV. PRESENTATIONS

"UV Fluorescence by Optical Pumping with Line Radiation," J.Trebes, Plasma Physics Colloquium, Yale, Nov.15, 1982.

"UV Fluorescence in CII pumped by AlIII line radiation",J.Trebes, NRL, Dec.3, 1982.

"Resonant Photo-excitation Lasers", M.Krishnan, Univ. of Rochester, Jan.26, 1983.

"Optically Pumped Short Wavelength Lasers," M.Krishnan, Atomic Physics Seminar, Yale Univ., Feb.1, 1984

"UV Fluorescence by Optical Pumping with Line Radiation," Poster Session, 2nd Topical Meeting on Laser Techniques in the Extreme Ultraviolet, Boulder, Colorado, March 5-7, 1984.

"Proposed new class of optically pumped, quasi-cw, uv and xuv lasers in the Be isoelectronic sequence," Poster Session, 2nd Topical Meeting on Laser Techniques in the Extreme Ultraviolet, Boulder, CO, March 5-7, 1984.

"Photo-excited, Be-like lasers", M.Krishnan, Lawrence Livermore Labs, CA, Jun.20,1984

"Short Wavelength Lasers-Research and Applications," Summer Research Program, Yale Univ.,Jul.11,1984

"Proposed new lasers in the Be-isoelectronic sequence-preliminary results in the uv," PPPL, Sep.11,1984

"Short Wavelength Lasers," M.Krishnan, Physics International Co. Oct.4,1984

"Design considerations for optically pumped, uv to xuv lasers in Be-like ions," Poster Session, APS-DPP Meeting,Boston MA, Oct.29-Nov.4,1984

"Observation of enhanced fluorescence at uv wavelengths in CIII

by optical pumping with MnVI line radiation," Poster Session, APS-DPP Meeting, Boston, MA, Oct.29-Nov.4,1984

"Optical pumping of CIII ions in a magnetically confined C plasma using MnVI line radiation from an adjacent Mn plasma," Poster Session, APS-DPP, Boston,MA, Oct.29-Nov.4,1984

"Photo-excited, extreme ultraviolet lasers," Institute for advanced study, (CTA), Sao Jose dos Campos, Brazil,Nov. 24,1984

"Photo-excited lasers from the uv to the xuv", M.Krishnan, Colloquium, Mechanical Engineering Dept., Univ. of Rochester, Jan.17, 1985

"Fluorescence in CIII by pumping with MnVI line radiation", M.Krishnan, invited paper at GEC, Monterey, CA, Oct 14,1985

"Fluorescence in CIII pumped by resonant line radiation from MnVI",M.Krishnan, invited paper at the Topical conference on the radiative properties of hot, dense plasmas, Williamsburg, VA, Oct.16,1985

"UV Fluorescence in CIII pumped by MnVI line radiation", Poster Session at APS-DPP Meeting, San Diego, Nov.4th,1985

"UV Fluorescence in CIII pumped by MnVI line radiation", N.Qi, Poster Session at NATO Advanced Study Institute on Radiative Processes in Discharge Plasmas, Pitlochry, Scotland, June 23-July 5,1985

"Fluorescence and small-signal gain in CIII by resonant photoex-citation with MnVI line radiation", N.Qi, Poster presented at

International Colloquium on X-ray lasers, Aussois, France, 14-17 April, 1986.

V. PUBLICATIONS

J.Trebes and M.Krishnan, "Ultraviolet fluorescence by optical pumping with EUV line radiation," Phys.Rev.Lett.50,679 (1983)

J.Trebes and M.Krishnan, "Observation of UV enhanced fluorescence due to the combined effects of selective optical pumping and collisional transfer," IEEE J. Quant. Electr. QE-19, 1870(1983)

M.Krishnan and J.Trebes, "Proposed new class of optically pumped, quasi-cw, uv and xuv lasers in Be-like ions," Appl. Phys. Lett.45,189 (1984)

M.Krishnan and J.Trebes, "Design considerations for optically pumped, quasi-cw, uv and xuv lasers in the Be-isoelectronic sequence," Laser Techniques in the extreme ultraviolet, (OSA, Boulder,CO,1984), S.E.Harris and T.B.Lcatorto, eds. (AIP,New York, 1984) p.514

J.Trebes and M.Krishnan, "UV fluorescence by optical pumping with line radiation," Laser Techniques in the extreme ultraviolet, (OSA, Boulder, CO, 1984), S.E.Harris and T.B. Lucatorto,eds, (AIP, New York, 1984) p.387

N.Qi, H.Kilic, and M.Krishnan, "Observed enhanced fluorescence at 2177, 2163 ,1923 and 1620Å in CIII using 310Å line radiation from MnVI," Appl. Phys. Lett. 46,471 (1985)

N.Qi, H.Kilic, and M.Krishnan, "Fluorescence and small signal gain

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at 2177 and 2163Å in CIII by resonant photo-excitation with 310Å, MnVI line radiation,"Journal de Physique, Aug. 1986

N.Qi and M.Krishnan, "Laser oscillation at 2177 and 2163Å in CIII, resonantly photo-excited by 310Å, MnVI line radiation," Phys. Rev. Lett. (to be published)

N.Qi and M.Krishnan,"Resonant photo-excitation lasers in Be-like ions," in preparation for Phys. Rev. A

N.Qi, R.R.Prasad, W.L.Morgan and M.Krishnan, "Ionisation balance and collisional-radiative kinetics in CII and CIII ions in vacuum-arc discharges," in preparation for J.Appl. Phys.

VI. SCIENTIFIC PERSONNEL

J.TREBES, graduate student, Ph.D, 1984

H-L.LU, graduate student, summer, 1982

C-J.LIU, graduate student, summer, 1982

R.R.PRASAD, graduate student, summer, 1982

N.QI, graduate student, Ph.D, 1986

H.KILIC, Associate Research Scientist, 1984-86

VII. ACKNOWLEDGEMENTS

UV FLUORESCENCE BY OPTICAL PUMPING WITH LINE RADIATION

James Trebes and Mahadevan Krishnan
Yale University, New Haven, Connecticut 06520

ABSTRACT

Optical pumping of CII ions in a vacuum arc discharge using AlIII ions in a laser produced plasma is described. The CII, 2p-5d, 560.437 Å transition was selectively pumped by line radiation from the AlIII, 3p-5s transition at 560.433 Å. The wavelength mismatch is less than the Doppler width of the AlIII line. Four transitions in CII, from the 5d, 5f, 4s, and $2p^2$ levels were studied simultaneously to examine the collisional-radiative redistribution of the pumped, 5d population. Electron density and temperature were measured in the C plasma. The Al plasma was characterized by measurements and numerical modeling in order to estimate the intensity of the AlIII pump line. A collisional-radiative model of the CII level populations was constructed with the measured density and temperature as inputs. Comparison of this model with the measurements allows discussion of the feasibility of building a UV laser with such a pumping scheme.

INTRODUCTION

Among the many approaches to the production of short wavelength population inversions is that of optical pumping with line radiation. In this approach, intense line radiation in one ion species is used to pump a nearly coincident transition from the ground state to a highly excited state in another ion species. The pumped, upper level may then be inverted with respect to lower lying levels. A survey of prior research in this field is given in a companion paper in these proceedings.[1] Recently, the work of Hagelstein[2] has motivated an experimental program[3] to test the feasibility of pumping soft X-ray lasers with such a scheme. Trebes and Krishnan[4,5] have demonstrated UV fluorescence by the combined effects of optical pumping and collisional transfer. This paper presents experimental results of the simultaneous measurement of enhanced fluorescence on four different transitions in CII, corresponding to four distinct upper states, when only one of these states was optically pumped with AlIII line radiation. Also presented are measurements and estimates of electron density and temperature in the C plasma as well as the Al pump plasma. These measurements enabled the development of a multi-level, collisional-radiative model for CII. The experimental observations are discussed in light of the model. Prospects for building a UV laser are discussed.

FLUORESCENCE MEASUREMENTS

This section begins by describing the experimental apparatus and characterizing the carbon discharge plasma. Then the measurements of fluorescence are described.

Figure 1 is a schematic diagram of the experimental apparatus. The carbon plasma is produced in a laser initiated vacuum arc, between the negative carbon cathode and a grounded carbon anode as shown. Two CO_2 TEA lasers are used. Laser I is focused on the cathode and triggers a vacuum arc discharge. The power supply for this discharge is a pulse-forming network with an external, impedance matching resistor. A typical oscillogram of discharge current vs time is shown in Fig. 2a. The flat-topped current duration is about 60 μs. Figures 2b, 2c, 2d, and 2e show typical line radiation vs time from lines of CI, CII, CIII, and CIV at wavelengths indicated on the figures. CI radiation is present only during the rising portion of the current pulse and after decay of the pulse. The CII and CIII radiation exhibits quasi-steady behavior, but the CIV intensity is seen to decrease during the latter portion of the discharge, although the current is constant. Such a decrease may be due to a decrease in electron temperature in the constant current arc, which in turn may be caused by dynamic effects. Based on these observations, it was decided to attempt optical pumping of the C plasma at a time of 40-45 μs after arc initiation, when quasi-steady conditions were observed.

The aluminum pump plasma was produced by focusing Laser II after the selected delay of typically 43 μs, onto an Al rod target, shown in Fig. 1. To ensure reproducibility, the Al rod was replaced after every ten laser shots. With each new target, five shots were fired to clean the target surface and then data were obtained with the next 5 shots. Earlier experiments[4,5] had shown that the 5d level in CII was pumped by AlIII, 560.433 Å line radiation from the adjacent Al laser produced plasma. Furthermore, the optical excitation was shown to be collisionally transferred to the 5f, higher angular momentum level. Enhanced fluorescence was measured on the 5d-3p and 5f-3d transitions at 2138 and 2993 Å, respectively. The primary motivation for the experiments described in this paper was to unravel the collisional-radiative kinetics in CII, following selective optical pumping. Toward this end, four different wavelengths in CII were monitored simultaneously. The wavelengths selected are shown in Fig. 3. The 2138 Å and 2993 Å lines were expected to show enhanced fluorescence due to optical pumping as before. The 3920 Å line would show fluorescence only if the 4s upper state were strongly coupled by collisions and radiative transitions to the 5d level. The 1335 Å line stems from an n = 2 level which is much lower in energy than the 5d and 4s levels. Furthermore, the $2p^2$ upper state of this line is not accessible by dipole, single electron transitions from the n = 5 or n = 4 shells. Therefore no fluorescence was expected at the
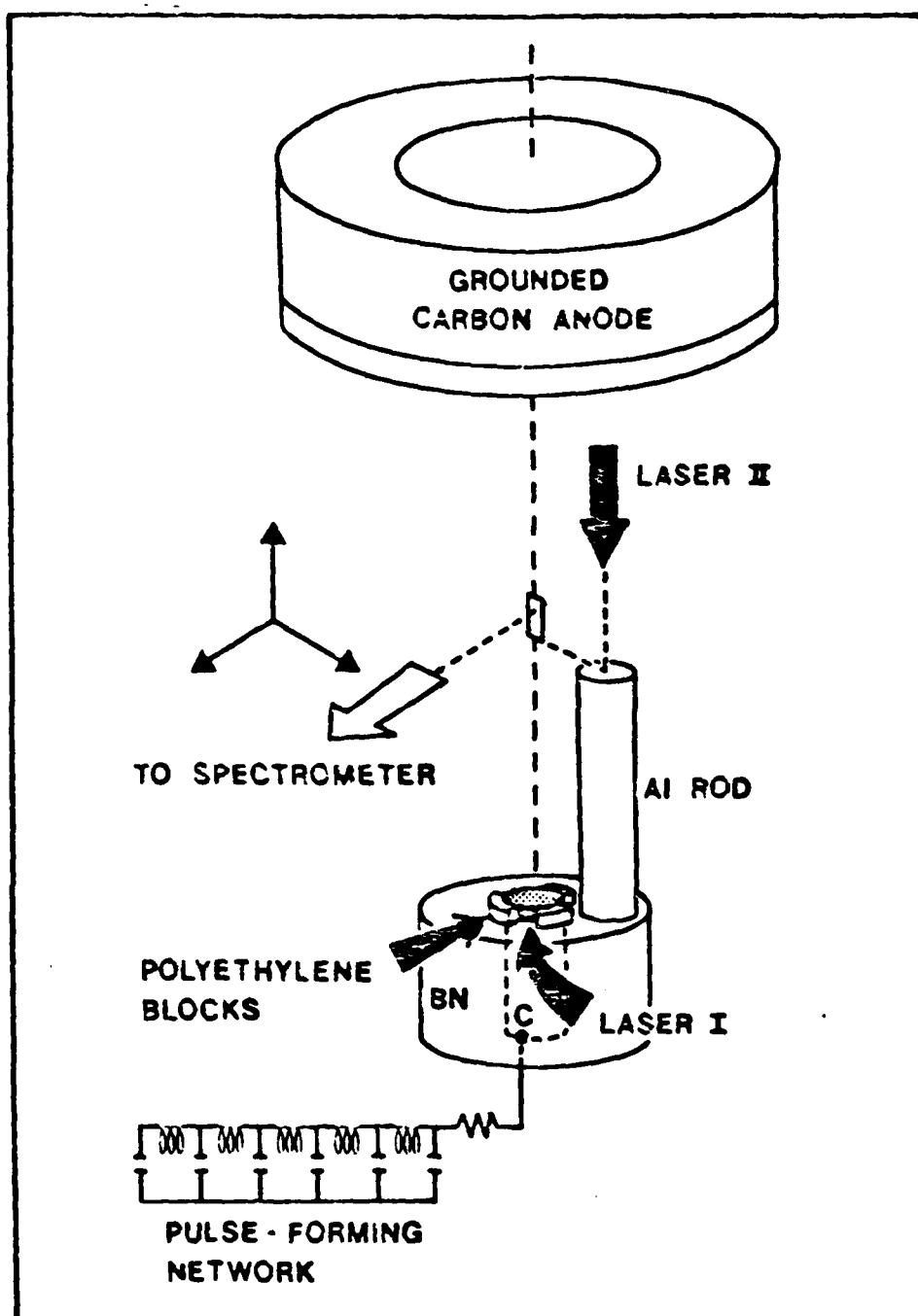
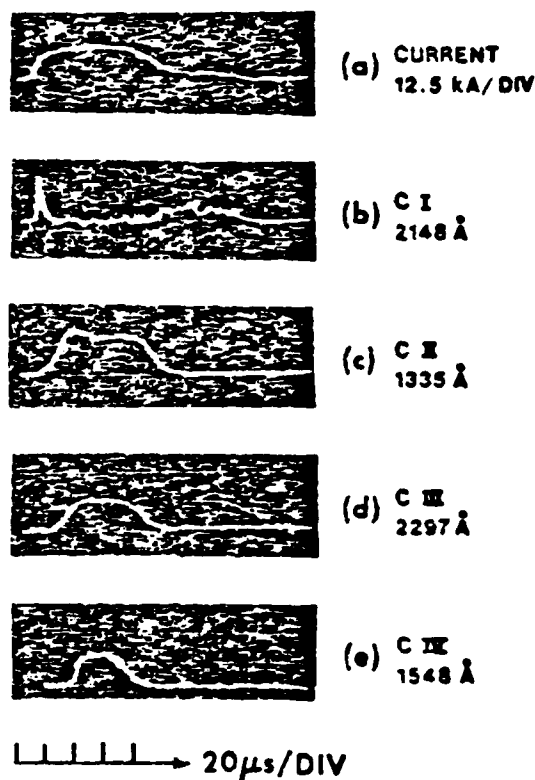Fig. 1.   Schematic diagram of the experimental apparatus.

Fig. 2. Discharge current and carbon line emission vs time in the
laser-initiated vacuum arc: a) Current 12.5 kA/DIV, b)
CI, 2148 Å line intensity, c) CII, 1335 Å line intensity,
d) CIII, 2297 Å line intensity, and e) CIV, 1548 Å line
intensity. The time scale is 20 μs/DIV.

1335 Å wavelength.

Figure 4 shows a Grotrian diagram of AℓIII. A .25 m Jarrel-
Ash monochromator was used to monitor the AℓIII, 3713 Å transition
in the laser produced Aℓ plasma. This wavelength was chosen be-
cause direct measurement of the 5p-3s, 560 Å pump transition was
hampered by inadequate resolution of the XUV spectrometer, as dis-
cussed later. It can be shown that the 5s upper state of the
3713 Å line and the 5p state are strongly coupled by collisions
in an expanding, laser produced plasma.[6] Therefore the intensity
of the 3713 Å line does provide a measure of the duration and rel-
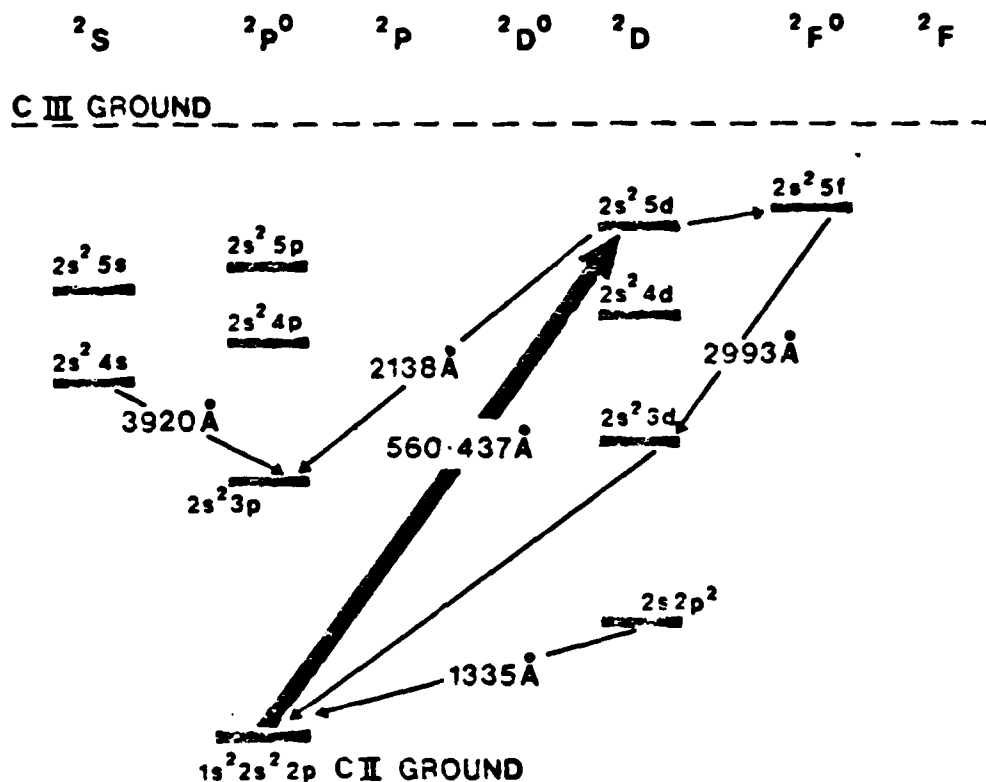ative intensity of the 560 Å pump line.

Fig. 3. Partial Grotrian diagram of CII, showing the wavelengths studied.

Figure 5 shows the arrangement of the spectrometers used to monitor the five wavelengths discussed above. All spectrometers, with the exception of the Jarrel-Ash, were focused onto the same local plasma region, 15 mm downstream of the cathode, along the discharge axis. The Jarrel-Ash was focused to a region 2 mm downstream of the Aℓ disc along the laser plasma axis.

In the first series of experiments, a fixed discharge current of 3.4 kA was arbitrarily chosen and the Aℓ plasma was produced 43 μs after discharge initiation. Ideally, the electron temperature in the carbon plasma should be such that the ground state population of CII is high, while the excited state population of the 5d level is low. Since it was difficult to directly measure the CII ground state population, the optimum discharge configuration was not established.

The observed fluorescence is shown in Fig. 6. Figure 6a shows three traces. The upper trace shows the CII, 2138 Å intensity, the middle trace the CII, 2993 Å intensity, and the lower trace the AℓIII, 3713 Å intensity vs time. Significant enhanced
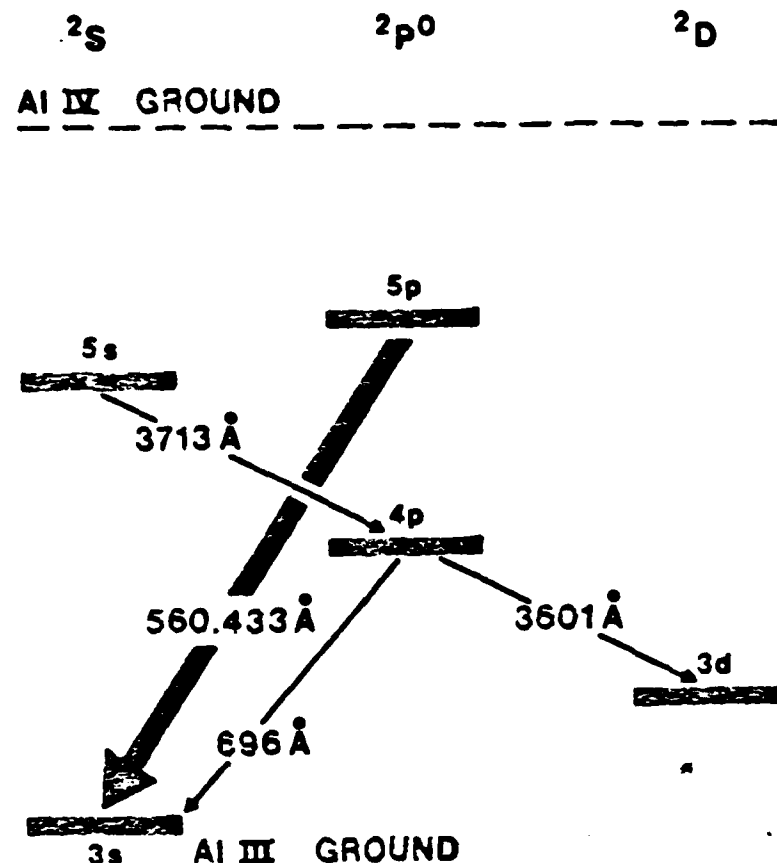
^{2}S $\qquad$ ^{2}P^0 $\qquad$ ^{2}D

Al IV GROUND ___ ___ ___ ___ ___ ___ ___

5p

5s

3713 Å

4p

560.433 Å

3601 Å

3d

696 Å

3s   Al III   GROUND

Fig. 4. Partial Grotrian diagram of AℓIII.

fluorescence is observed on both CII lines, coincident with the Aℓ plasma. Figure 6b shows the CII, 3920 Å intensity (upper trace) and the CII, 1335 Å intensity (middle trace) vs time. The lower trace is again the AℓIII, 3713 Å intensity. It is observed that both of these CII wavelengths also show enhanced fluorescence due to the Aℓ plasma. To ensure that the observed fluorescence was not due to continuum or to the wings of neighboring Aℓ lines from the Aℓ plasma leaking into the spectrometers, the experiments were repeated with no carbon discharge and only the Aℓ laser produced plasma. The results are shown in Figs. 6c and 6d. Some spurious background does appear, but the observed fluorescence in Figs. 6a and 6b is larger than this background for all four wavelengths.

The observed fluorescence at 1335 Å was puzzling, since the $2p^2$ upper state is not directly coupled to the upper levels. However, this state is strongly coupled to the CII ground state by a dipole allowed transition. It was possible that electrons from the denser and hotter Aℓ plasma collisionally excited this transition. This conjecture was supported by the different time scale
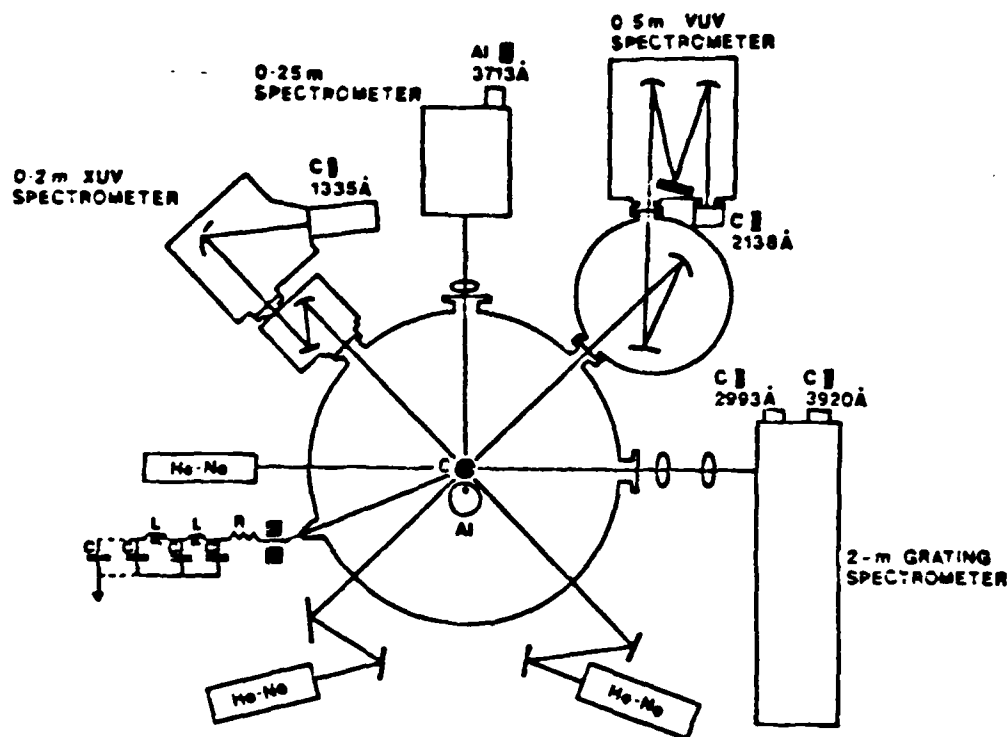
Fig. 5. Schematic diagram of the spectrometer arrangement.

for the 1335 Å fluorescence as compared with that for each of the
other three wavelengths. Figure 7 shows the data of Fig. 6 on two
timescales, 20 μs/DIV and 2 μs/DIV. The 1335 Å fluorescence is
seen to decay more rapidly than the other lines. To test this con-
jecture, the Aℓ rod target was replaced with a Mg rod and the ex-
periments were repeated. Since no lines of Mg are coincident with
any CII transitions, no optical pumping was expected. Figure 8
shows the results. Figure 8a shows the time evolution of the CII,
2138, 2993, and AℓIII, 3713 Å line radiation. Figure 8b shows
the CII, 3920, 1335, and AℓIII, 3713 Å lines. As with Fig. 6, the
background measurements are shown in Figs. 8c and 8d. It is seen
from the figures that although there appears to be some enhanced
fluorescence coincident with a Mg pump plasma, the apparent en-
hancements on all but the 1335 Å line are in fact due to spurious
background. The rather large background at 2138 Å is probably due
to the wings of a bright, MgIII line at 2135 Å. The 1335 Å line
shows enhanced fluorescence clearly above the background, suggest-
ing that for both the Aℓ and Mg plasmas, this transition was prob-
ably excited by electrons penetrating the C discharge from the
laser produced plasma. Further work is needed to confirm this.
Nevertheless, these results serve to underscore the point made in
earlier papers[4],[5] that optical pumping with line radiation is best
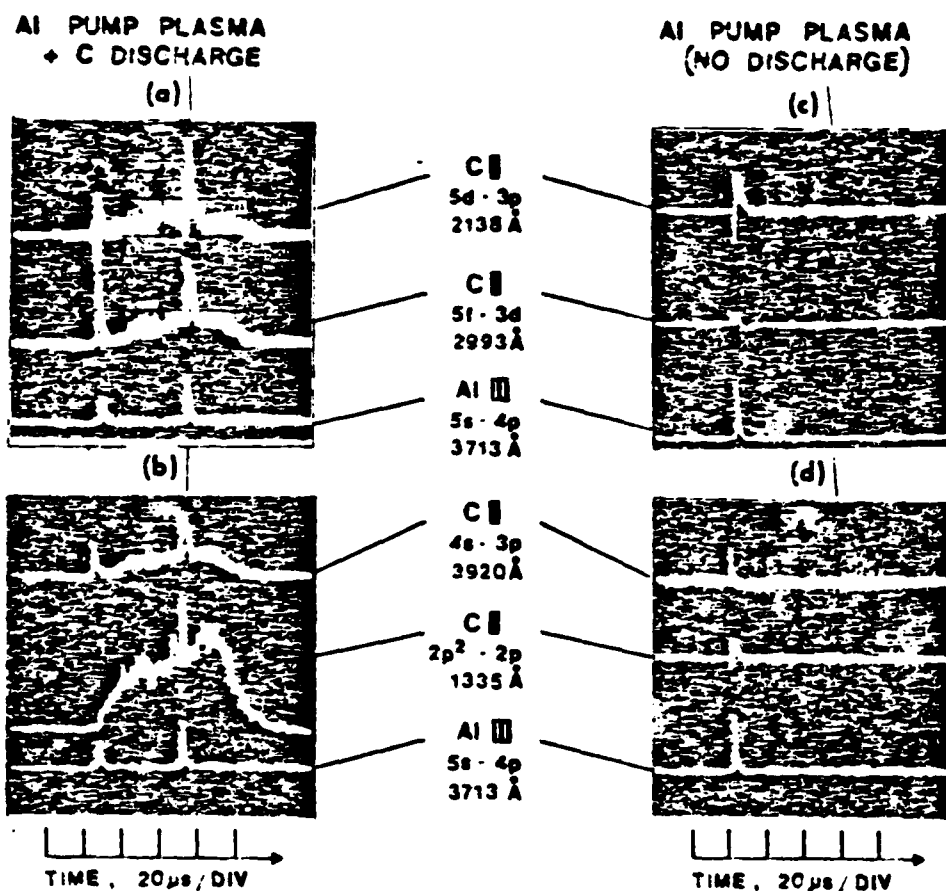achieved by using two separate plasmas which are well isolated

Fig. 6. Optically pumped fluorescence in CII: a) intensity vs time of CII, 2135 and 2993 Å lines, and AℓIII, 3713 Å line; b) intensity vs time of CII, 3920 and 1335 Å lines, and AℓIII, 3713 Å line. 6c) and d) same lines as in 6a and b, but with no C discharge to show spurious background signals due to Aℓ laser produced plasma alone.

from each other.

The preceding discussion has shown that in the carbon discharge, selective optical pumping to the 5d level is accompanied by strong coupling of the pumped level to other levels in the $n = 5$ and $n = 4$ shells. It is possible that the $n = 3$ levels are also coupled to higher levels. Such coupling is not conducive to producing population inversions at the 2138 and 2993 Å UV wavelengths. A detailed, collisional-radiative model must be constructed to examine such coupling. Necessary inputs to such a model are the electron density n_e and electron temperature T_e in the carbon plasma. Experiments were performed to measure these parameters. The results are described in the next two sections.
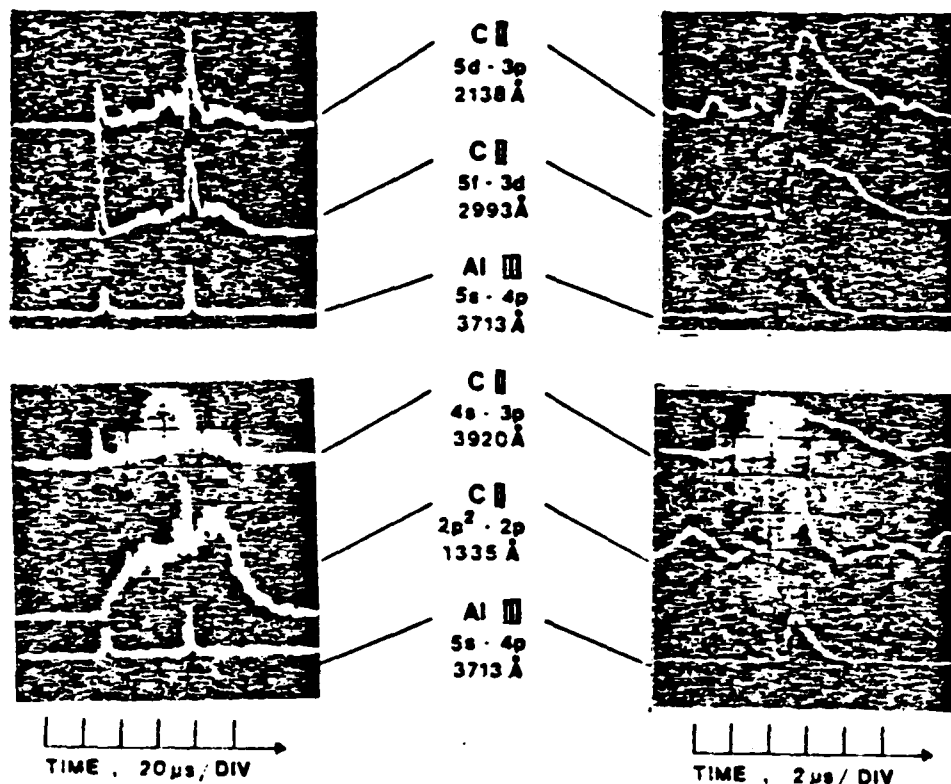
AI PUMP PLASMA
+ C DISCHARGE

TIME , 20 μs/DIV

TIME , 2 μs/DIV

Fig. 7. Data of Fig. 6a and 6b displayed on two timescales of
20 μs/DIV and 2 μs/DIV, respectively.

ELECTRON DENSITY MEASUREMENT

Electron density in a laser-initiated carbon vacuum arc simi-
lar to that used in these experiments was measured by Keren and
Hirshfield,[7] by measuring the refraction of a far-infrared laser
beam after propagation through the plasma. For a cathode identical
to that used here, but with the vacuum vessel walls serving as
anode, n_e was measured for discharge currents I from 2 to 8 kA.
The measurements were fit to a power law dependence on current,
viz.:

$$n_e = 3 \times 10^{14} \ I^{1.9} \ \cos\phi/r^{2.4} \tag{1}$$

where: r is the radius in cm and ϕ is the angle from the discharge
axis. For a radius of 1.5 cm in these experiments, the above for-
mula gives a density on axis of 1.2×10^{15} cm^{-3}, for a current of

Mg PUMP PLASMA
+ C DISCHARGE (a)

Mg PUMP PLASMA
(NO DISCHARGE) (c)

(b)

(d)

C II
5d-3p
2138Å

C II
5f-3d
2993Å

Mg
CONTINUUM

C II
4s-3p
3920Å

C II
2p²-2p
1335Å

Mg
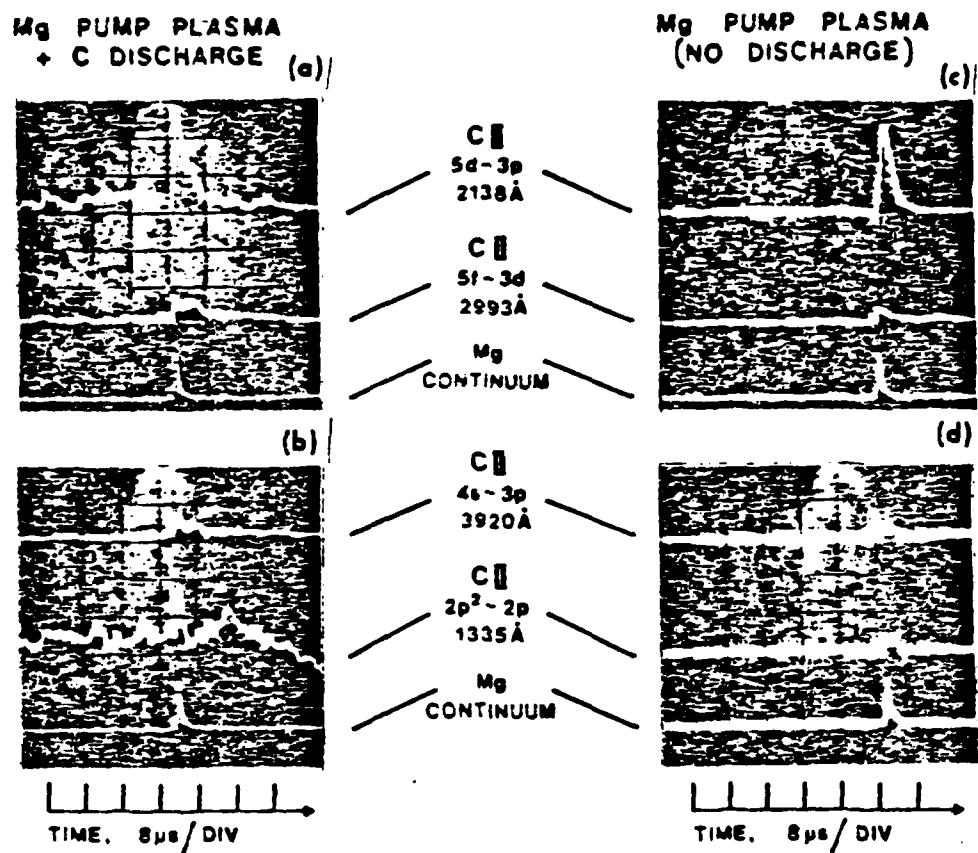CONTINUUM

TIME, 8μs/DIV

TIME, 8μs/DIV

Fig. 8. C discharge with an adjacent Mg laser produced plasma:
a) intensity vs time of CII, 2138 and 2993 Å lines, and
AℓIII, 3713 Å line; b) intensity vs time of CII, 3920
and 1335 Å lines, and AℓIII, 3713 Å line. 8c) and d)
same lines as in a and b, but with no C discharge, to
show spurious background signals due to Mg laser produced
plasma alone.

3.4 kA. To corroborate this estimate, hydrogen atoms were intro-
duced into the arc and the Stark width of the H_β line was meas-
ured. The hydrogen was introduced by arranging three segments of
polyethylene on the surface of the boron nitride insulator
(Fig. 1). The 2 m Ebert spectrometer was used for these measure-
ments, with a resolution of 0.2 Å. The Stark profile was measured
by scanning a photomultiplier across the focal plane at 0.2 Å in-
tervals. Ten shots were fired at each wavelength and the average
line intensity was recorded. To ensure reproducibility of the
data, the Ly_α, 1216 Å line intensity and the CII, 1335 Å line in-
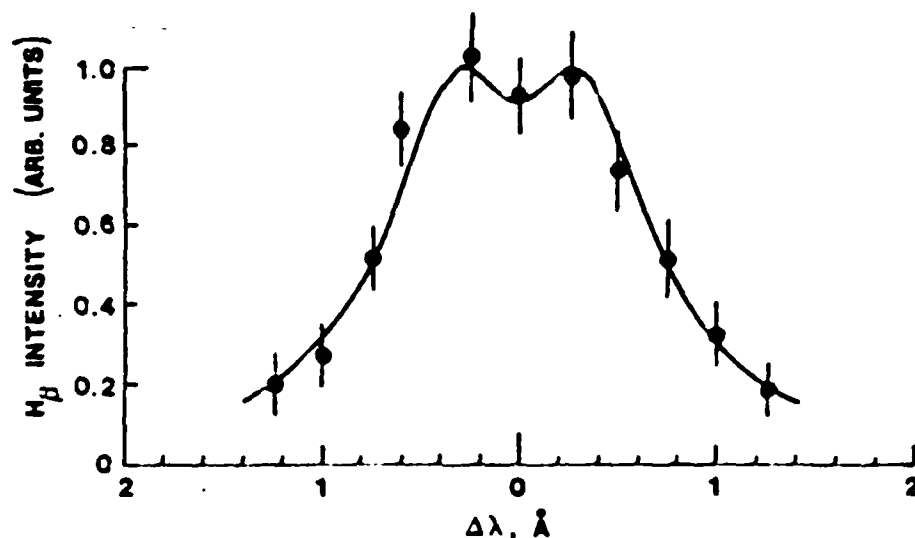tensity were monitored simultaneously with the H_β signal. Repro-

Fig. 9. H_β line intensity vs wavelength in the C discharge. The curve through the measured points is a convolution of a Stark profile with a Doppler profile.

ducible data were obtained by cleaning the entire cathode assembly after every 30 shots. The measured H_β line intensities at 3.4 kA, at 50 μs after arc initiation are shown in Fig. 9. The curve through these points is a best fit of a Stark profile,[8] convoluted with a Doppler profile. The density obtained is 6×10^{14} cm^{-3}, for a best fit temperature of 0.4 eV. The dip at the center of the Stark profile is a sensitive measure of temperature, because of the convolution of the Doppler profile. For example, a higher assumed temperature of 1 eV would completely wash out the dip. In the next section it will be shown that the temperature in the C arc on the axis is about 3 eV. Modified coronal calculations show that the H_β intensity is very sensitive to temperature.[6] It would appear therefore that the measured H_β signals originated predominantly from outer regions of the arc, where the temperature is lower. The measured density of 6×10^{14} cm^{-3} is thus a lower bound and consistent with the earlier estimate of 1.2×10^{15} cm^{-3}, from the scaling law of Keren and Hirshfield.[7]

ELECTRON TEMPERATURE MEASUREMENT

At densities of $\sim 1 \times 10^{15}$ cm^{-3} and temperatures of ~ 3 eV, it can be shown[6] that high lying levels of CI and CII are in thermodynamic equilibrium with the ground levels of CII and CIII, respectively. Measurement of the relative intensities of transitions from such higher levels in CI and CII thus leads to a deter-

mination of the ratio of the ground level populations of CII and
CIII, as a function of temperature. Independently, a modified
coronal equilibrium calculation also yields the ratio of the
ground level populations of CII and CIII, as a function of temp-
erature. The intersection of these two independent functions then
yields the electron temperature. The transitions chosen were at
2478 Å in CI and 2993 Å in CII. The measured intensity ratio of
2993/2478 was 27, at 50 μs after discharge initiation. This ratio
corresponds to a CII/CIII ground state ratio of 1, at a tempera-
ture of 3.25 eV. Figure 10 shows the relative abundance of the
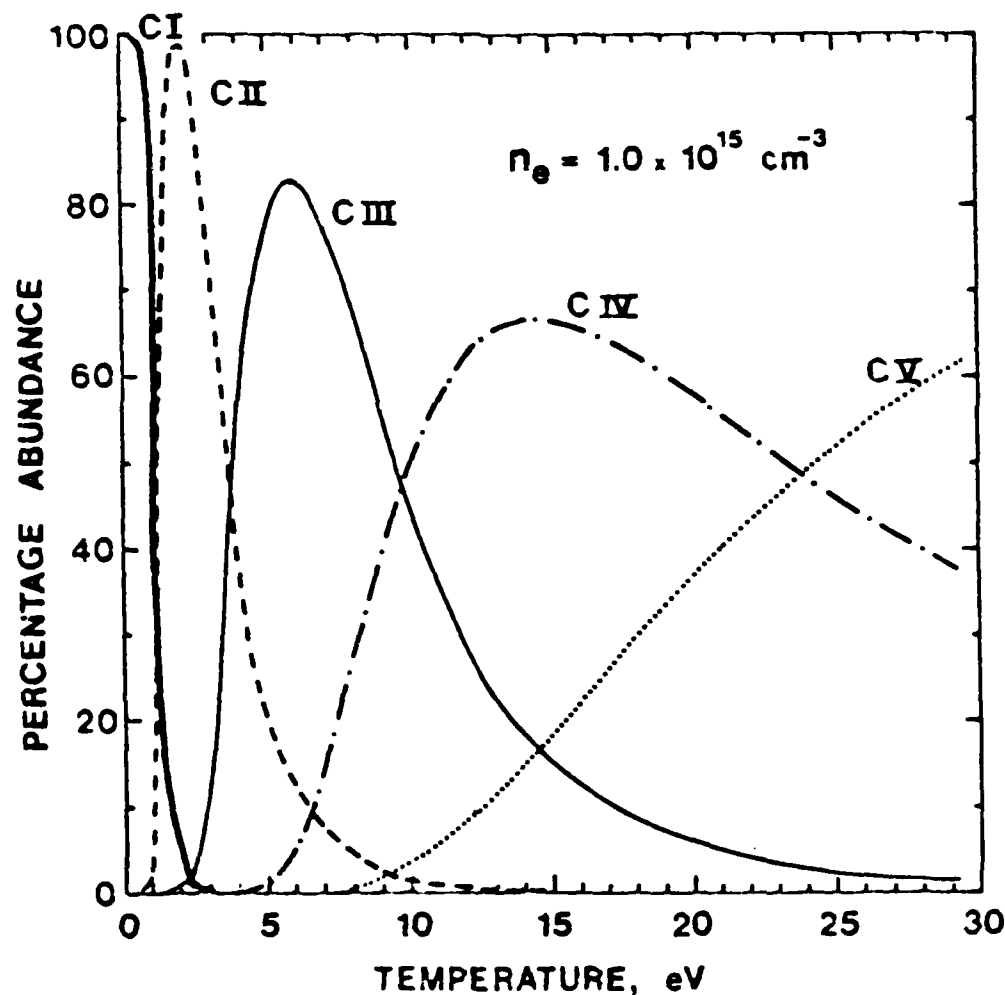different carbon charge states vs temperature, as derived from the

Fig. 10. Relative abundance of C charge states vs temperature,
obtained from a modified coronal equilibrium calcula-
tion. $n_e = 1 \times 10^{15}$ cm^{-3}.

modified coronal equilibrium model.[1] The coronal equilibrium also gives a CII/CIII ground state ratio of 1 at the same temperature of 3.25 eV. The uncertainty in the measurement translates to a temperature uncertainty of ±0.25 eV. At the measured temperature of 3.25 eV, Fig. 10 shows that there is a negligible fraction of CIV in the plasma. At 3.4 kA, the resonance line of CIV at 1548 Å could not be detected.

To recapitulate, the carbon discharge plasma was found to have an electron density of 1×10^{15} cm^{-3} and an electron temperature of 3.25 ± 0.25 eV. Under these conditions, the n = 5 levels of CII are in thermodynamic equilibrium with the CIII ground state, thus causing the optical excitation of the 5d level to be rapidly distributed by collisions over a large number of higher levels. Population inversions and lasing on the 5d-3p and 5f-3d transitions are therefore difficult to achieve. In the next section, the Aℓ pump plasma is examined with a view to estimating the 560 Å pump line intensity.

PUMP LINE INTENSITY

In these experiments, it was not possible to measure directly the density and temperature of the laser produced plasma or to measure the absolute intensity of the pump transition. However, some measurements were made which together with numerical modeling allowed a reasonable estimate to be made of the 560 Å pump radiation. The initial temperature of the Aℓ plasma was determined by using an empirical relation[9] based on the laser focal spot intensity on the Aℓ target. The temperature was found to be ~10 eV. The initial density at a distance of 0.1 mm downstream of the target surface was estimated as 5×10^{18} cm^{-3}, based on measurements of Tonon and Rabeau[10] in a similar plasma. At this density and temperature, the modified coronal model[1] predicts that most of the ions will be in charge states higher than AℓIII. The AℓIII must therefore be formed in the expansion phase of the laser produced plasma. This expansion leads to a rapid decrease in density and temperature, such that "freezing" of the populations occurs, with the resultant charge state distribution characterized by an effective temperature which is much higher than the local electron temperature. Such a non-equilibrium expansion makes it hard to interpret spectroscopic measurements in the expansion phase of the plasma.

One approach to determining the temperature was by measuring the relative intensity of the 3713 Å, 5s-4p and 3601 Å, 4p-3d transitions in AℓIII (see Fig. 4). If the collisional coupling rates between the 5s and 4p levels greatly exceed the radiative rate, then the two levels are in collisional equilibrium and the 3713/3601 line ratio yields an excitation temperature for AℓIII. It is shown[6] that this is the case for $n_e > 1 \times 10^{16}$ cm^{-3}. The excitation temperatures obtained as a function of time at various axial positions downstream of the target surface are plotted in Fig. 11. Curiously, the temperature appears to increase well
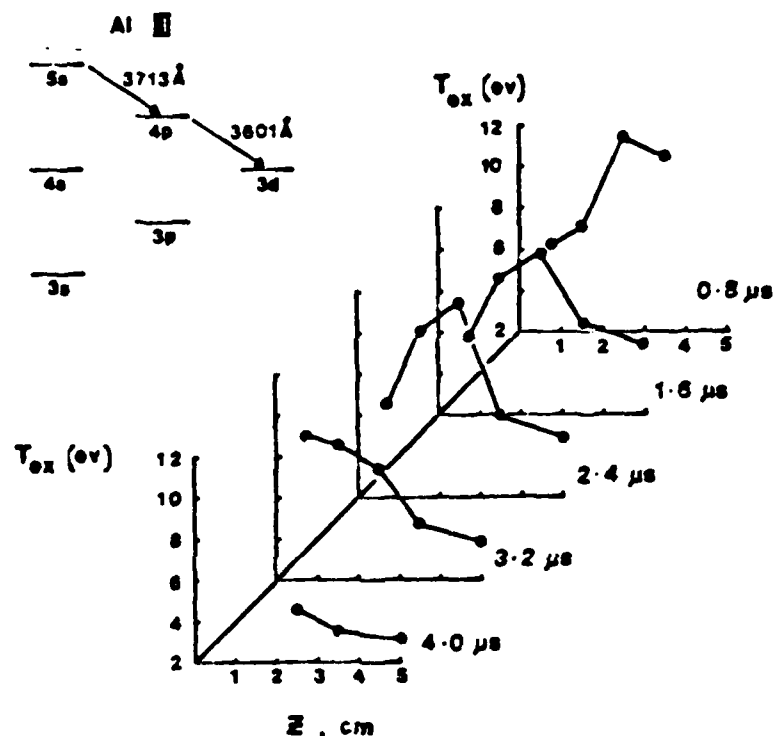
EXCITATION TEMPERATURE IN Al LASER PRODUCED
PLASMA FROM INTENSITY RATIO OF $3601Å/3713Å$
Al III LINES

Fig. 11.   Excitation temperature derived from the AlIII
           3713/3601 Å line ratio plotted as a function of
           time and of distance downstream from the Al target.

after the laser pulse, reaches a maximum and then decreases. This
anomalous behavior persists for up to 3 µs at distances up to 3 cm
downstream. The cause of this anomaly is optical trapping of the
3601 Å radiatior. which causes an increase in intensity of the
3601 Å line and results in an anomalously low temperature derived
from the line ratio. At greater distances downstream and for
later times in the expansion, the plasma density is sufficiently
low that the 3601 Å line is optically thin and the derived temp-
erature decreases with time as expected. The density of the 3d
level required to give an optical depth of unity at 3601 Å is
found to be 1×10^{13} cm^{-3}, for an ion temperature of 1 eV. With
this lower bound, and assuming that the excitation temperature is
11 eV (based on the line ratios), the lower bound on the 5s and
5p densities is found to be 1×10^{12} cm^{-3}. For distances less
than 3 cm and times shorter than 3 µs, the densities are clearly
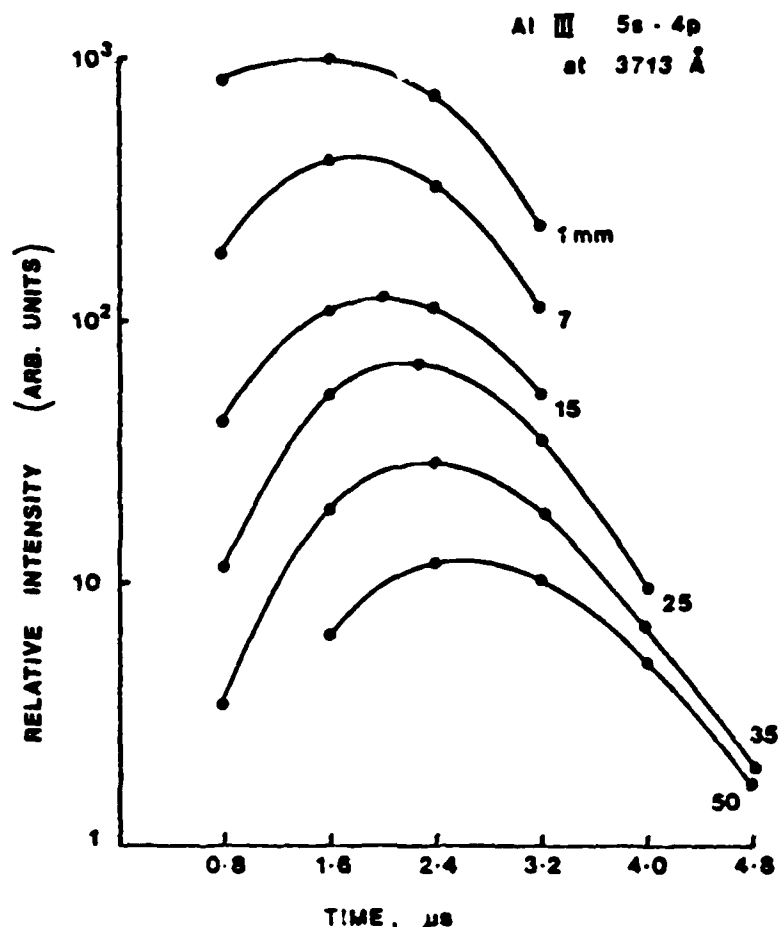higher. The persistence of intense, AlIII radiation for such long

Al III   5s · 4p
at   3713 Å

Fig. 12. AℓIII, 3713 Å line intensity vs time, for several
          axial positions downstream of the Aℓ target, along
          the target normal.

times in the plasma expansion is further verified in Fig. 12,
which shows the AℓIII, 3713 Å intensity vs time, for several posi-
tions downstream. The intensity is highest for distances less
than 3 cm at times less than 3 μs. The above detailed description
of the AℓIII, 3713 Å line intensity indicates the presence of
excited states of AℓIII in the rapidly expanding laser produced
plasma. Direct measurement of the AℓIII, 560 Å line shape was not
possible because the 1.5 Å resolution of the XUV spectrometer was
about 150 times larger than the estimated Doppler width of the
560 Å line. Even to detect this line, therefore, the line inten-
sity must be 150 times higher than that of the continuum back-
ground. Time-resolved intensity measurements of the 560 Å as well
as the AℓII, 4p-3s, 696 Å line are shown in Fig. 13. Figure 13a
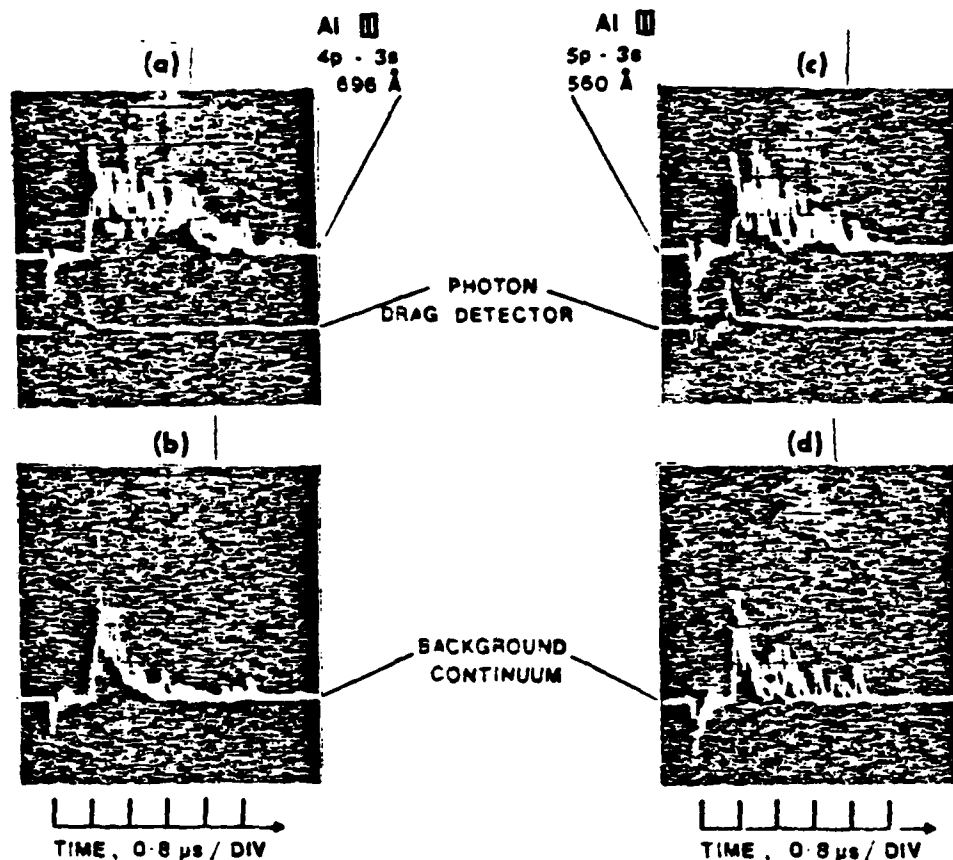shows the 696 Å intensity vs time, while Fig. 13b shows the back-

Fig. 13. Time-resolved AℓIII line intensities in laser produced
plasma: a) AℓIII, 4p-3s, 696 Å intensity vs time;
b) continuum background; c) AℓIII, 5p-3s, 560 Å inten-
sity vs time; d) continuum background. The lower
trace in a and c is the signal from the photon drag
detector which samples the CO_2 laser.

ground continuum, measured 5 Å from line center. Figure 13c shows
the 560 Å intensity vs time, and Fig. 13d shows the background
continuum, 5 Å from line center. In each case, the figure shows a
superposition of three laser shots on the photograph. The lower
trace on Figs. 13a and 13c is the output of a photon drag detector
which samples a portion of the CO_2 laser beam. Both 696 Å and
560 Å line radiation signals emerge above the continuum background
for about 3 μs. This time duration is consistent with the obser-
vation of enhanced fluorescence also for 3 μs. In conclusion, the
lack of an absolutely calibrated XUV spectrometer as well as the
non-equilibrium expansion of the laser plasma made it impossible
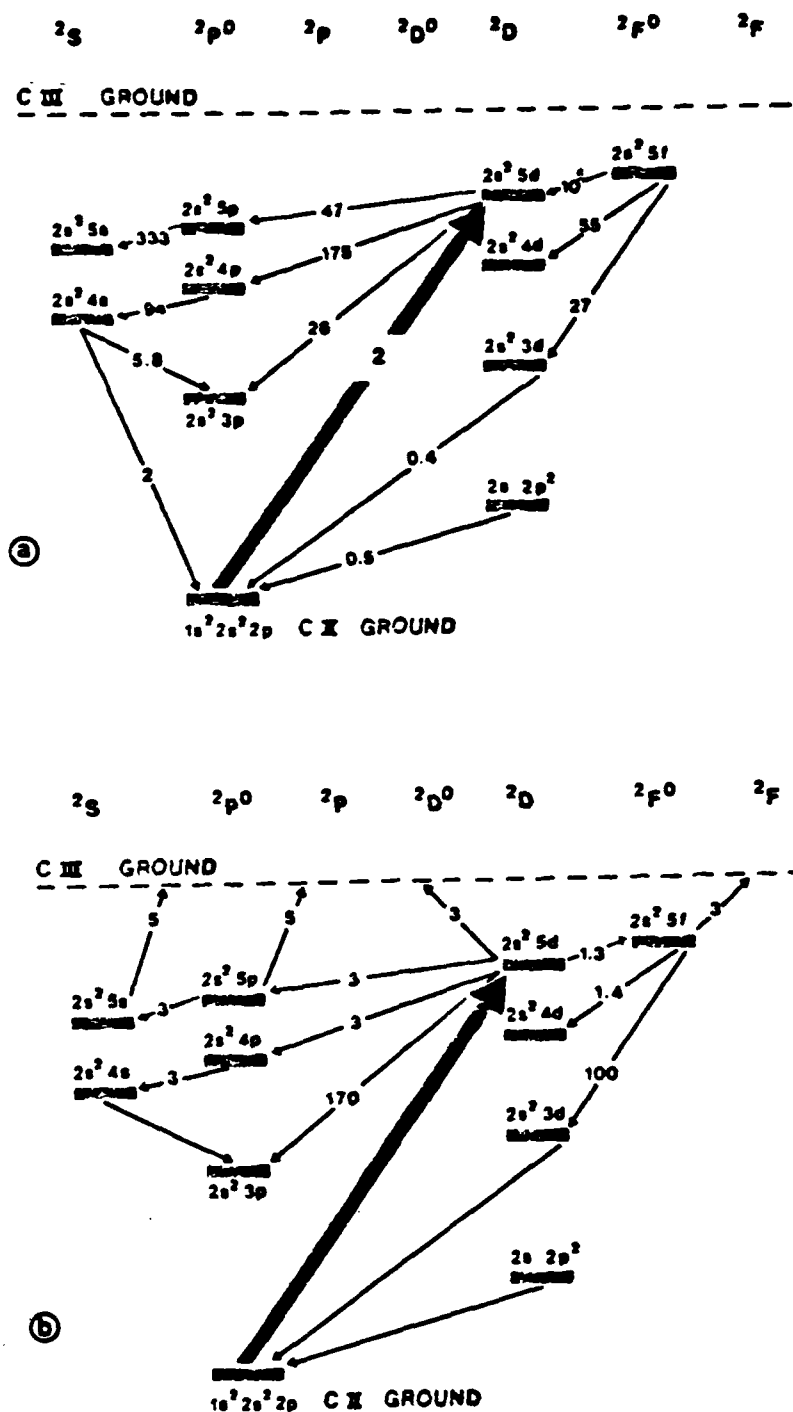to accurately estimate the pump line intensity.

Fig. 14. Radiative and collisional times for transitions in CII.
a) Radiative lifetimes (in ns) and b) collisional
ionization and de-excitation times (in ns), for selected
transitions in CII.

high n_e and T_e in the carbon plasma is that collisional rates are high enough to tend to thermalize the n = 3, 4, and 5 shells in CII. Figure 14b shows the collisional ionization rates for the n = 5 levels, the collisional transfer rates between these levels, and some de-excitation rates for the transitions considered in Fig. 14a. From the figure, it is observed that the collisional transfer time from 5d to 5f is short compared to the 5d radiative lifetime. This is desirable in order to transfer the pumped electrons in the 5d to the 5f, potential upper laser level. However, the figure also shows that collisional ionization from the 5d and 5f levels proceeds as rapidly as the collisional transfer. Furthermore, the collisional de-excitation times for the 5d and the 5f levels are an order of magnitude or more shorter than the radiative lifetimes. This means that the optically pumped 5d population is rapidly distributed by collisions to the other levels in the n = 4 and 5 shells, as well as to higher lying levels and the CIII ground state. It is clear that conditions in the carbon plasma are not optimal for a 5f-3d laser, since selective optical pumping of the 5d level is not accompanied by collisional transfer exclusively to the 5f level. Another conclusion that may be drawn from the above analysis of collisional rates is that for a given strength of the optical pumping, enhanced fluorescence should be seen simultaneously on several transitions from the high lying levels of CII, since the collisional coupling times between levels are orders of magnitued shorter than the 3 µs duration of optical pumping. This conclusion is verified by the observed fluorescence at 3920 Å from the 4s level, discussed earlier.

The above discussion has revealed much of the kinetics of the CII ions without regard to the actual strength of the optical pump. Using the estimate of the lower bound of the AℓIII, 5p population of 1×10^{12} cm^{-3}, it is possible to estimate the degree of enhanced fluorescence expected from the pumped 5d level. With the CII ground state density of 6.7×10^{14} cm^{-3}, convolution of the AℓIII and CII Doppler line shapes and inclusion of the solid angle subtended by the observed volume to the Aℓ plasma yields[6] an expression for the lower bound of the optical pump rate:

$$\text{Pump Rate} = 9.7 \times 10^{11} \frac{1}{\sqrt{T_{A\ell} T_C}} L_{A\ell} \times \Delta\Omega \times n_{5p} \times A_{5p-3s} \quad I' \quad (2)$$

where $T_{A\ell}$ and T_C are the ion temperatures of Aℓ and C, respectively, $L_{A\ell}$ is the effective Aℓ plasma dimension, $\Delta\Omega$ the solid angle subtended by the pumped carbon volume to the Aℓ plasma, n_{5p} is the lower bound estimate of the AℓIII, 5p population, A_{5p-3s} is the 560 Å transition probability and I' is a numerical estimate of the convolution integral of the two Doppler line shapes. For the conditions discussed throughout this paper, the lower bound of the pump rate is determined to be 10^{18} electrons/cm^3 s. For the 5d level, the sum of all possible collisional and radiative de-exci-

To summarize the experimental measurements, optical pumping of CII with AℓIII line radiation was studied by examining simultaneously four different wavelengths in CII. Enhanced fluorescence was observed from the pumped 5d level as well as from the neighboring 5f level. In addition, fluorescence was also observed from the 4s and $2p^2$ levels. The 4s level was probably fed by collisions from the n = 5 shell whereas the $2p^2$ level was probably collisionally excited from the CII ground state by electrons from the laser plasma. The electron density in the C arc was measured to be 1.5×10^{15} cm^{-3}. The electron temperature was estimated to be 3.3 eV. The AℓIII, 560 Å pump line radiation was found to persist for up to 3 μs in the laser produced plasma. In the next section, the collisional-radiative kinetics of the optically pumped CII ions are examined. The experimental observations are compared with the model and the feasibility of pumping a laser using such a scheme is discussed.

COLLISIONAL-RADIATIVE KINETICS IN CII

Using the measured density and temperature as inputs, a collisional-radiative model was developed for CII. This model, described in detail in Trebes,[6] included for a given level: collisional excitation and de-excitation, collisional ionization, radiative and three-body recombination from the CIII ground state, and dipole allowed radiative transitions. The energy levels and some oscillator strengths were obtained from standard references.[11,12] Other oscillator strengths and various rate coefficients were obtained from Morgan.[13] The model developed allows calculation of the distribution of the populations in many levels, subsequent to the selective optical pumping of the 5d level. Some results are shown in Fig. 14. Figure 14a shows the energy levels, some allowed transitions and their radiative lifetimes in ns. From the radiative lifetimes indicated in Fig. 14a, it appears that the 5f-3d transition at 2993 Å is a potential candidate for a quasi-cw laser, since the 5f radiative lifetime is much longer than the 3d radiative lifetime. However, one major drawback is the strong coupling of the 3d lower level with the CII ground state, which can lead to optical trapping of the 3d-2p, 800 Å radiation. From the modified coronal model, the CII ground state density was estimated to be 6.7×10^{14} cm^{-3}, for the measured n_e and T_e. For this ground state density, the optical depth at line center of the transition is 27.2, for an assumed transverse plasma dimension of 1 cm. Using the Holstein escape factor,[14] the modified lifetime of the 3d level is 1ℓ ns. Since this modified lifetime is still shorter than the radiative lifetime of the 5f level, quasi-cw lasing is possible, but if the transverse plasma dimension is higher or if the CII ground state density is slightly higher, then optical trapping increases the lifetime of the 3d level to a value higher than the 5f lifetime, thus destroying the possibility of a quasi-cw laser at 2993 Å. A second deleterious consequence of the rather

tation rates as well as the collisional ionization rate is 10^{10} s^{-1}. Thus the steady state population enhancement of the 5d level by optical pumping is 10^8 cm^{-3} or higher. In the absence of optical pumping, the 5d population is estimated from the collisional-radiative model to also be about 10^8 cm^{-3}. These estimates imply that the enhanced fluorescence should be comparable to the spontaneous emission from the 5d level. If the pump line intensity is up to a factor of ten higher than the estimated lower bound, the enhanced fluorescence should then be a factor of ten above the spontaneous emission. In the earlier measurements[4,5] and in this work, the observed fluorescence was always between one and ten times the spontaneous emission, in agreement with the predictions of the model. In addition, the model predicts that the degree of enhancement at the 2993 Å wavelength should be lower than that at 2138 Å, since only a fraction of the enhanced 5d population is transferred to the 5f level. This is also in agreement with the experimental observations.

To summarize, a detailed examination of the collisional-radiative kinetics in CII has revealed that the density and temperature in the carbon plasma are far from ideal for creating a population inversion. Collisions dominate the kinetics and cause strong coupling between the n = 3, 4, and 5 levels, as well as higher levels. Thus, although the optical pumping is selective, the pumped population is dispersed over many channels.

CONCLUSION

A detailed experimental and theoretical study has been made of optical pumping in CII ions using AℓIII line radiation. This study has shown that although the optical pumping itself is selective, the pumped 5d population in CII is dispersed into many competing channels by collisional and radiative processes. Also, the strong collisional coupling between levels in the n = 3, 4, and 5 shells renders it difficult to sustain an inversion between 5f and 3d, a potential laser transition. Optical trapping also increases the 3d lifetime and further contributes to spoiling the chance for an inversion. These deleterious effects may be alleviated somewhat by a better choice of density and temperature for the carbon plasma. For example, as n_e is reduced, the collisional rates for various decay channels out of the 5d level are all proportionally reduced. For $n_e = 10^{13}$ cm^{-3}, the 5d-5f electron collisional transfer time is 300 ns, which is still short compared to the optical pumping duration of 3 µs. For transitions within a given shell, particularly when the energy gap is very small compared to the temperature, the ion collisional transfer rate can sometimes exceed the electron collisional rate. For the 5d-5f transition, ion collisions are more important than electron collisions.[6] Thus the collisional transfer time is even less than 300 ns. At this n_e, 5d-3p radiative decay is ten times as rapid as collisional ionization or collisional de-excitation. If the collisional transfer time from 5d-5f is comparable with the 5d-3p radiative

decay, up to 50% of the optically pumped 5d population may be transferred to 5f. Also, at this lower n_e, the CII ground state density is lower, and optical trapping of the 3d level is avoided. Finally, the 5f-3d radiative decay is then five times as rapid as collisional de-excitation via 5f-4d. Therefore a population inversion may be produced between 5f and 3d. It is important to point out that although the CII ground state density is only 6.7×10^{12} cm^{-3}, if only 10% of the ground state is optically pumped to the 5d level, then the 5f population would be about 3.0×10^{11} cm^{-3}. At 2993 Å, this corresponds to a small signal gain of .1 cm^{-1}. If the gain medium is 10 cm in length, the resultant net gain is quite sufficient to sustain oscillation in a cavity.

In retrospect, the CII-AℓIII combination was chosen because of the good coincidence between the pump and absorption line wavelengths. In these experiments, the plasma conditions were not optimized for lasing. The CII plasma was too dense and hot, while the AℓIII pump species were produced in the non-equilibrium expansion of a laser produced plasma under conditions far from ideal for maximizing the pump line intensity. Nevertheless, some enhanced fluorescence was observed, with only 1×10^{-6} of the CII ground state being pumped to the 5d level. Under optimized conditions, 10% or more of the ground state population may be pumped to the 5d level and a very high gain laser is possible. These arguments are tempered by the observation that the major stumbling block of this particular ion combination is the unfavorable energy level structure of the CII ion. Firstly, the pumped 5d level is too close to the CIII ground state and thus readily ionized. Secondly, the n = 4 levels are close to the n = 5 levels and strongly coupled to them by superelastic collisions. Finally, since the 3d lower level of the potential laser transition is directly coupled to the ground state, optical trapping is a serious concern. A better approach to producing a laser using optical pumping with line radiation would consider both the line coincidences as well as the atomic level structure of the pumped ion. Just such considerations have led to the proposal of a new class of optically pumped lasers in Be-like ions. These schemes are described in a companion paper in these proceedings.[1]

ACKNOWLEDGMENTS

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Observed enhanced fluorescence at 2177, 2163, 1923, and 1620 Å in C III by photoexcitation with Mn VI line radiation at 310 Å

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Line radiation at 310.182 Å from Mn VI ions in a laser produced plasma was used to resonantly pump C III ions in an adjacent, vacuum arc discharge from the $2s$ ^1S ground level to the $4p$ $^1P^0$ upper level. Enhanced fluorescence by up to a factor of 150 was measured on the $4p$ $^1P^0$–$3d$ ^1D line at 2177 Å. Collisional exchanges between the $n = 4$ levels transfer the pumped $4p$ population to the $4d$ and $4f$ levels. Enhanced fluorescence was also measured on the $4d$ ^1D–$3p$ $^1P^0$ line at 1620 Å and on the $4f$–$3d$ lines at 2163 Å and 1923 Å, respectively.

In 1961, Rabinowitz, Jacobs, and Gould[1] reported laser oscillation at 7.12 μ in Cs I, when the upper laser level was photoexcited by 3880-Å resonance line radiation from a helium lamp. Djeu and Burnham[2] extended this concept of selective photoexcitation to a visible wavelength by pumping a 5461-Å laser in Hg I in a vapor cell, using line radiation from Hg I in an adjacent cell. Recently, Krishnan and Trebes[3] have proposed a new class of Be-like, photoexcited lasers with wavelengths from 2177 Å in C III to 213 Å in Mg IX. This letter reports observed fluorescence from the $4p$, $4d$, and $4f$ levels in C III, when the $4p$ level was pumped by 310-Å line radiation from Mn VI. The radiative lifetime ratio of $4p/3d$ is 40, while that for $4f/3d$ is 7.5. Both the $4p$-$3d$ and the $4f$-$3d$ lines are thus candidates for quasi-cw, ultraviolet lasers. Selective photoexcitation with line radiation has also been proposed as a means for pumping soft x-ray lasers in H-like and He-like ions.[4,5] Extensive numerical analyses[5–7] suggest that terawatt lasers and terawatt level pulsed-power technology are capable of providing adequate pump power in line radiation with which single-pass, high-gain lasers may be produced at soft x-ray wavelengths. At ultraviolet wavelengths, the pump power required is orders of magnitude lower, and smaller gains are tolerable because of the availability of high reflectance optics. The results presented in this letter were obtained with a pump laser power of 100 MW. Since C III is isoelectronic with Mg IX, the collisional-radiative kinetics elucidated by this research shed light directly on possible soft x-ray lasers in Mg IX.

Figure 1 shows a schematic diagram of the experimental apparatus. The C III ions are produced in a laser triggered vacuum arc discharge between a 6-mm-diam carbon cathode and a hollow carbon anode 100 mm downstream. The discharge, from a pulse-forming network, with typically 5-kA current and 60-μs pulse duration, is triggered by focusing laser I onto the cathode. At a distance of 15 mm downstream from the cathode, the electron density and temperature are 10^{15} cm^{-3} and 3 eV,[8] respectively. About 40 μs after discharge initiation, the pump plasma is produced by focusing laser II onto a Mn target on a rotatable disc as shown in Fig. 1. Lasers I and II are both 15 J/100 MW, CO_2 lasers. The focal spot of laser II is 7 mm off axis from a point 15 mm downstream from the cathode. Spontaneous emission and enhanced fluorescence in C III were measured by a vacuum ultraviolet spectrometer which imaged a 0.1×2 mm region on the discharge axis at the 15-mm downstream location.

A partial energy level diagram of C III and C IV is shown in Fig. 2. The $2s$ ^1S–$4p$ $^1P^0$ transition at 310.17 Å is resonantly pumped by 310.182-Å line radiation from Mn VI ions in the laser produced plasma. The Doppler width of the Mn VI line at a temperature of 30 eV is 17 mÅ, which is greater than the 12-mÅ wavelength mismatch. Photoexcitation of C III by Mn VI was therefore expected, with enhanced fluorescence on the $4p$ $^1P^0$–$3d$ ^1D line at 2177 Å. At $n_e \sim 10^{15}$ cm^{-3} and $T_e \sim 3$ eV in the C plasma, electron and ion collisions will rapidly transfer the $4p$ population to the $4s$, $4d$, and $4f$ levels.[7] Enhanced fluorescence was therefore also expected from these levels. Typical results obtained at 2177 Å are shown in Fig. 3.

Figure 3(a) shows the C discharge current versus time. Figure 3(b) shows the spontaneous emission at 2177 Å, in the absence of the Mn pump plasma. When the Mn plasma was produced 43 μs after discharge initiation, the resultant enhanced fluorescence at 2177 Å is shown in Fig. 3(c). To capture this trace on film, the gain of the photomultiplier detector was kept identical to that for Fig. 3(b), but the sensitivity of the recording was reduced by a factor of 20. Hence the spontaneous emission is not visible above the baseline. To determine what fraction of the observed fluorescence at 2177
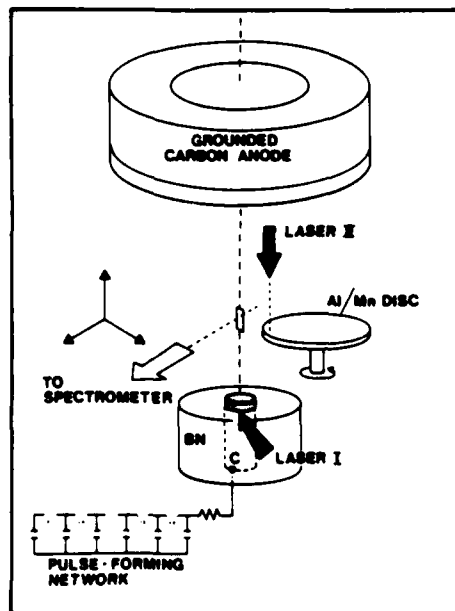

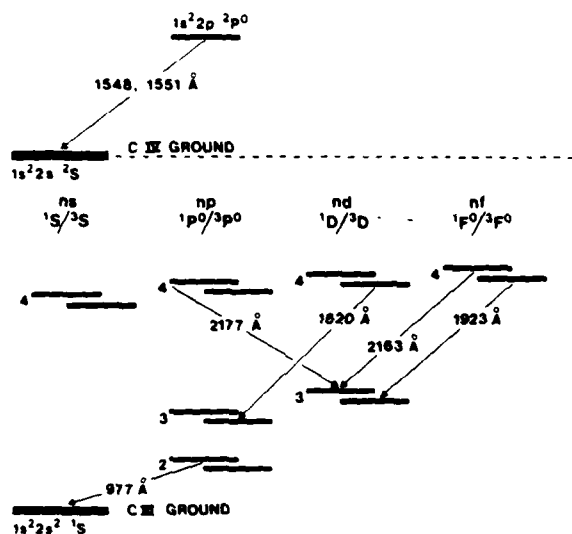FIG. 1. Schematic diagram of the experimental apparatus.

FIG. 2. Energy levels in C III and C IV. Optical pumping of the $2s^2$ ^1S–$2s4p$ $^1P^0$ transition is accompanied by collisional transfer to the $4p$ $^3P^0$, $4d$, and $4f$ levels. Quasi-cw lasing is possible on the $4p$-$3d$ and $4f$-$3d$ transitions.

Å might be due to line or continuum radiation from the Mn plasma which enters the field of view of the spectrometer, the C discharge was turned off and only the Mn plasma was produced. Figure 3(d) shows that under these conditions, there is indeed some spurious background detected at 2177 Å, but this is much less than the observed fluorescence from the C III ions. After subtracting this background from the total signal [Fig. 3(c)] and taking the scale factor into ac-
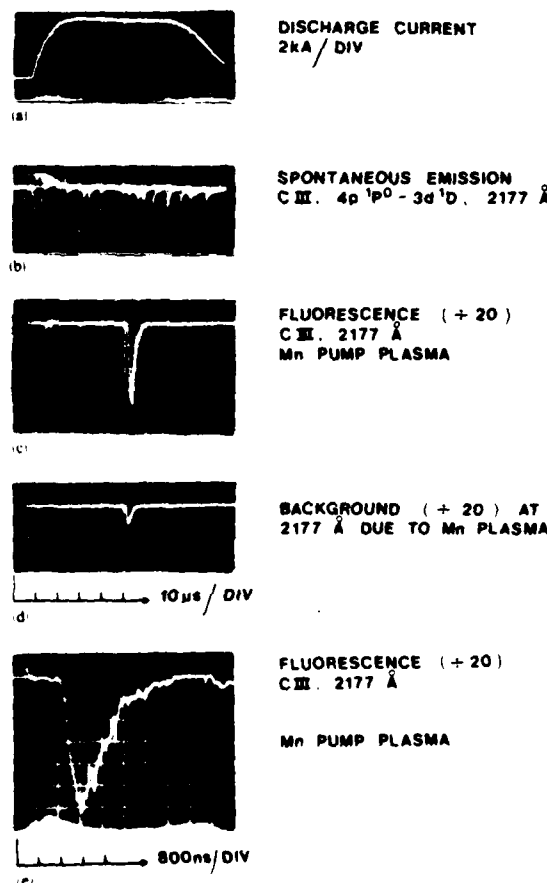

DISCHARGE CURRENT
2 kA / DIV

SPONTANEOUS EMISSION
C III. $4p$ $^1P^0$ - $3d$ ^1D, 2177 Å

FLUORESCENCE (+ 20)
C III. 2177 Å
Mn PUMP PLASMA

BACKGROUND (+ 20) AT
2177 Å DUE TO Mn PLASMA

10 µs / DIV

FLUORESCENCE (+ 20)
C III. 2177 Å

Mn PUMP PLASMA

800ns/DIV

FIG 3 Observed fluorescence at 2177 Å in C III.

count, the ratio of fluorescence/spontaneous emission at 2177 Å is between 100:1 and 150:1. Figure 3(c) actually shows data from three consecutive shots superimposed, and reveals the good reproducibility of the fluorescence. Figure 3(e) shows the enhanced fluorescence with better temporal resolution. The fluorescence is observed to persist for about 1.8 µs after the Mn plasma is produced. The radiative lifetime of $4p$ is 5 ns. Thus if the $4p$ level is inverted with respect to the $3d$ level, a quasi-cw laser at 2177 Å is possible. It might be argued that the observed fluorescence is due to collisional excitation of the $4p$ level in C III by hot electrons from the laser produced Mn plasma which expand into the C discharge. If this were the case, then a laser produced Al plasma should also produce enhanced fluorescence. When an Al pump plasma was produced at the same location as the Mn plasma, no enhanced fluorescence was observed. In addition to this test, the C II, $2s^2$ $2p$ $^2P^0$–$2s2p^2$ ^2D line at 1335 Å was also examined. Neither the Mn nor the Al plasma produced enhanced fluorescence at this wavelength, confirming that the fluorescence observed at 2177 Å was indeed due to selective photoexcitation by MnVI line radiation.

At the densities and temperatures typical of the carbon arc, it has been shown[7] that electron and ion collisions rapidly transfer the pumped $4p$ population to the other $n = 4$ levels. The $4d$-$3p$ and $4f$-$3d$ lines were therefore examined for enhanced fluorescence. At the $4d$ ^1D–$3p$ $^1P^0$ wavelength of 1620 Å, for a discharge current of 5.6 kA, the ratio of enhanced fluorescence to spontaneous emission observed was 30:1. The enhancement ratio decreased further, to 10:1 and 15:1 at the $4f$-$3d$ wavelengths of 2163 and 1923 Å, respectively. Figure 4 shows typical data at 1923 Å, corresponding to the $4f$ $^3F^0$–$3d$ ^3D line. Figure 4(a) shows the enhanced fluorescence when the Mn plasma was created 43 µs after initiation of the 5.6-kA C discharge. After subtraction of the spurious background (not shown), the enhancement ratio is about 15:1. Figure 4(b) shows the fluorescence when the discharge current was reduced to 3.3 kA. At this lower current, although the fluorescence remains the same, the spontaneous emission has decreased significantly, thus increasing the enhancement ratio.

FLUORESCENCE
C III. $4f$ $^3F^0$ - $3d$ ^3D. 1923 Å
Mn PUMP PLASMA

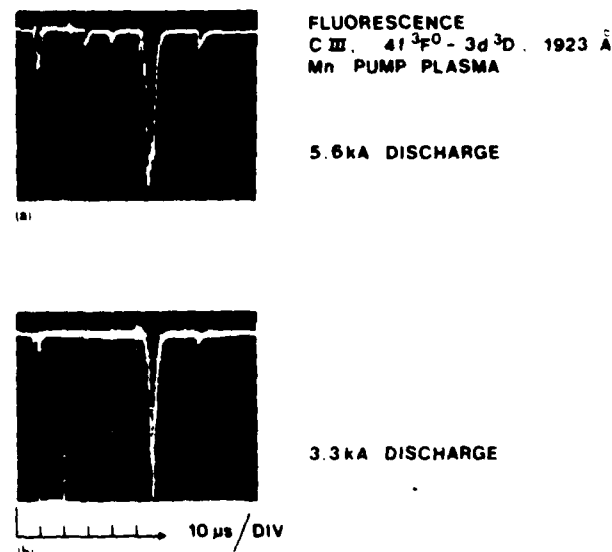5.6 kA DISCHARGE

3.3 kA DISCHARGE

10 µs/DIV

FIG. 4. Observed fluorescence at 1923 Å in C III.

For fixed pump plasma and discharge conditions, the fluorescence enhancement ratio decreases from about 150:1 at 2177 Å to 30:1 at 1620 Å and 15:1 at 2163 and 1923 Å. Earlier analysis[7] had shown that the $n = 4$ levels in C III should be driven into statistical equilibrium on the time scale for radiative decay from $4p$. Also, the times for single-step collisional ionization from the $n = 4$ levels were found to be much longer than the radiative lifetimes. When the $n = 4$ levels are in statistical equilibrium, the ratios of the $4s$, $4p$, $4d$, and $4f$ level populations should be 0.0625:0.1875:0.3625:0.4375. Hence most of the pumped $4p$ population should reside in $4d$ and $4f$ and the enhancement ratios for these levels should be as high as that for the $4p$ level, contrary to the experimental observations. There are two possible causes for this discrepancy: firstly, the C plasma density and the collisional rate coefficients may be lower than estimated, and secondly, the $4p$ level may be depopulated by multistep collisional ionization or by photoionization. A rough estimate of the photoionization rate is given by assuming that the Mn plasma radiates as a blackbody. For the photoionization rate to be comparable with the collisional rates within the $n = 4$ shell, the blackbody temperature required is about 20 eV. Since the Mn plasma in the initial expansion phase has such a temperature or higher, photoionization of the $4p$ level is possible. To test this notion, the spectrometer was tuned to the C IV, $2p\,^2P^0$–$2s\,^2S$ resonance line at 1551 Å. The upper state of this line is coupled to the C IV ground state by a strong dipole transition, and should therefore reveal an increase in the C IV ground state population. Under conditions identical to those described earlier,

the ratio of enhanced fluorescence/spontaneous emission measured at 1551 Å was about 12:1. Such an increase could also be due to stepwise collisional excitation from the $4p$ level. Further research is needed to clarify this deleterious process. One possible way to alleviate photoionization is by creating the Mn plasma in a vacuum spark discharge rather than by a CO_2 laser. The vacuum spark discharge may be tailored to produce a plasma consisting predominantly of Mn VI ions, at a density high enough to generate an intense, 310-Å pump line, but low enough to minimize broadband radiation. With the laser produced Mn plasma as a pump, we have already observed up to 150:1 enhancements in fluorescence at 2177 Å. Experiments are under way to optimize this fluorescence and to construct an oscillator at both this and the $4f$-$3d$ wavelengths

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FLUORESCENCE AND SMALL-SIGNAL GAIN AT 2177 AND 2163 Å IN CIII BY RESONANT PHOTOEXCITATION WITH 310 Å, MnVI LINE RADIATION

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Abstract - Line radiation at 310 Å from a laser-produced Mn plasma was used to resonantly photoexcite the $2s\ ^1S - 4p\ ^1P^0$ transition in CIII ions in a vacuum-arc discharge. Enhanced fluorescence, up to a factor of 180, was observed on the 4p-3d line at 2177 Å. Enhanced fluorescence was also observed on other 4-3 lines in CIII, because the photoexcited 4p population is collisionally redistributed among all the n = 4 levels. A 72-level, collisional-radiative model of CIII was constructed, including the resonant photoexcitation. Measured values of electron density and temperature were used in the model. Theoretical predictions of enhanced fluorescence agree well with the measurements. Small-signal gain coefficients of ~0.1 cm^{-1} were theoretically predicted. Single-pass gain measurements show gains as high as 0.4 cm^{-1} on the 4p-3d, 2177 Å line and the 4f-3d, 2163 Å line. This CIII-MnVI photoexcitation scheme is a prototype for soft x-ray lasers, using higher Z, isoelectronic analogs.

I. INTRODUCTION

There are many approaches to soft x-ray lasers. Population inversion mechanisms include direct collisional excitation, three-body or dielectronic recombination, broadband inner-shell photoexcitation, and resonant photoexcitation. Recently, high gain at 206 and 209 Å was demonstrated [1] in a Ne-like, Se laser-produced plasma. Inversion between the $2p^5\ 3p$ and $2p^5\ 3s$ levels in Ne-like Se appears to be driven by a combination of direct collisional excitation from the $2p^6$ ground state as well as dielectronic recombination from the F-like $2p^5$ ground state. The search for a three-body recombination pumped laser has produced noteworthy results [2] in CVI at 182 Å, in a laser-produced magnetically confined plasma. A third approach to soft x-ray lasers is by resonant photoexcitation of ions in one plasma by intense line radiation from ions in an adjacent plasma. This approach was proposed in 1975 by Vinogradov, Sobelman, and Yukov [3], and by Norton and Peacock [4]. Several other resonant photoexcitation schemes have been proposed [5] and analyzed [6], and some experiments [7] have been reported, in He-like ions. Krishnan and Trebes [8] proposed a class of photoexcited lasers in Be-like ions. Quasi-cw lasers with wavelengths from 2177 Å in CIII to 213 Å in MgIX were identified, in an isoelectronic sequence.

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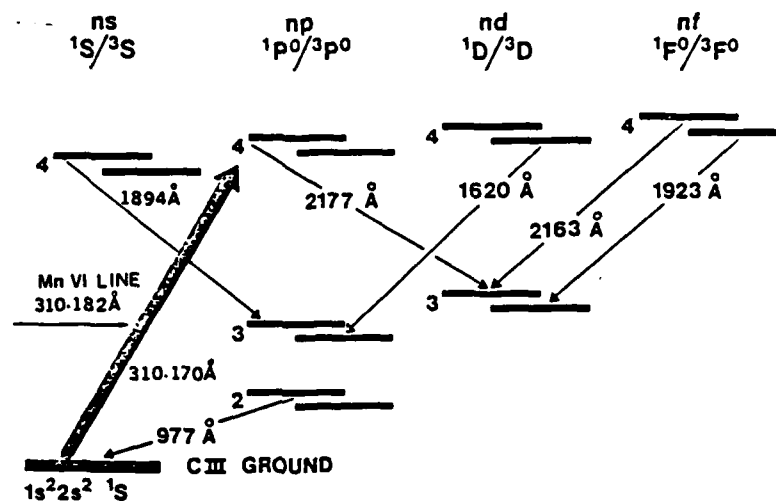
Fig. 1. Energy levels in CIII. Optical pumping of the $2s^2 \, ^1S$ - $2s4p \, ^1P^0$ transitions is accompanied by collisional transfer to the 4s, 4d, and 4f levels. Lasing is possible on the 4s-3p, 4p-3d, and 4f-3d transitions.

Figure 1 illustrates the principle of resonant photoexcitation. MnVI line radiation at 310.182 Å pumps the CIII ions from the $2s^2$ ground state to the $2s4p \, ^1P^0$ level. For a sufficiently strong Mn pump, inversion occurs between 4p and 3d, and a laser is possible at 2177 Å. Collisions in the CIII plasma may rapidly thermalize the n = 4 level populations. In this case, gain is also possible on other n = 4-3 transitions as shown in the figure. This CIII-MnVI laser scheme is a prototype for soft x-ray lasers in higher Z, isoelectronic analogs. Qi, Kilic, and Krishnan reported [9] enhanced fluorescence on several 4-3 transitions in CIII ions in a vacuum-arc discharge, pumped by MnVI line radiation from a Mn, laser-produced plasma.

This paper describes further experiments on the prototype CIII-MnVI scheme. Measured fluorescence due to photoexcitation is compared with the predictions of a 72-level, collisional-radiative model. This model for CIII was constructed using an atomic data base provided by Dr. W.L. Morgan of Lawrence Livermore Laboratory. The model predicted gains of ~0.1 cm^{-1} at 2177 Å, 2163 Å, and 1894 Å in CIII. Using an optimized discharge geometry, small-signal gains (~0.4 cm^{-1}) were measured, using a single-pass reflection technique.

II. FLUORESCENCE MEASUREMENTS

Figure 2 is a schematic drawing of the experimental arrangement. The CIII plasma is produced in a vacuum-arc discharge between a hollow carbon anode and a 6 mm diameter solid C cathode as shown. The interelectrode separation is 100 mm. The cathode is located 5 mm off the optical axis of a 0.5 m VUV spectrometer, which was used to measure fluorescence and gain on the n = 4-3 lines of CIII. The C cathode is biased negatively by up to -6 kV by a pulse-forming network. A vacuum-arc discharge is triggered by focusing a 15 J/150 MW CO_2 laser onto the cathode. A typical discharge current waveform is shown in Fig. 3(a). The rise-time is 15 μs, the flat-topped width about 50 μs, and the peak current is 6400 A. The discharge plasma so produced consists mostly of CIII ions, with an electron density n_e - 10^{16} cm^{-3} and temperature T_e ~ 4 eV. The Mn pump is produced by focusing another 15 J/150 MW CO_2 laser to a 2 mm x 23 mm line focus on a Mn slab as shown. The line focus is 7 mm off the spectrometer axis. Typically, the Mn laser-produced plasma is produced 36 μs after discharge initiation.

Typical enhanced fluorescence measurements are shown in Fig. 3. The horizontal time scale in the figures is 10 μs/div. Figure 3(b) shows the spontaneous emission on the 4p $^1P^0$ - 3d ^1D line at 2177 Å. The trace shown is an average over five successive discharges. Figure 3(c) shows the enhanced fluorescence at 2177 Å, when the
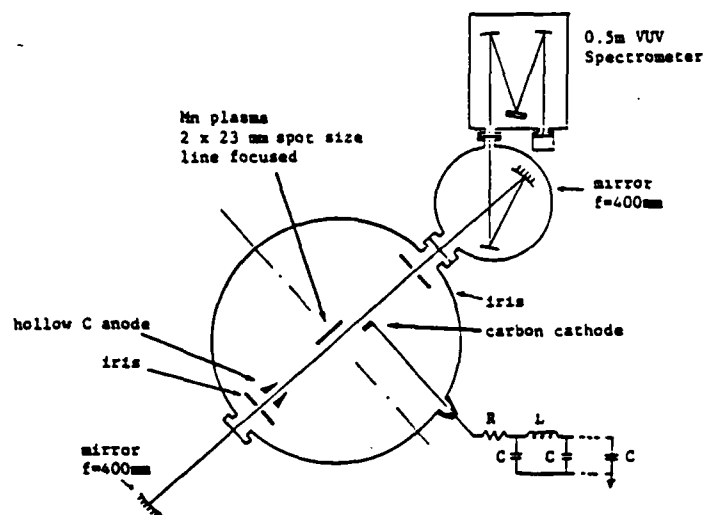
Fig. 2. Schematic diagram of the experimental apparatus.

a. DISCHARGE CURRENT
4.0KA/DIV

b. SPONTANEOUS EMISSION
CIII 2177Å

c. FLUORESCENCE (÷45)
CIII 2177Å

10μS/DIV
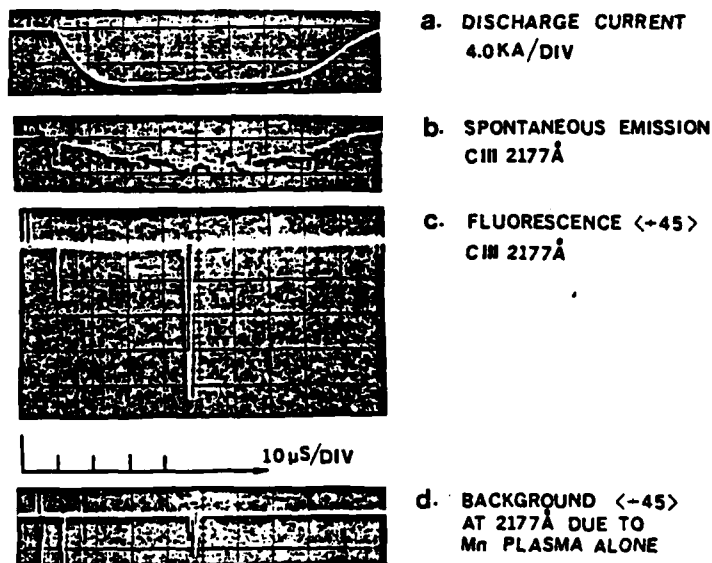
d. BACKGROUND (÷45)
AT 2177Å DUE TO
Mn PLASMA ALONE

Fig. 3. Typical observed fluorescence at 2177 Å in CIII. (a) Discharge current vs
time; (b) spontaneous emission vs time (no optical pumping); (c) enhanced
fluorescence at t = 36 μs with MnVI line radiation; (d) spurious background
due to Mn plasma alone.

Mn laser-produced plasma was created, 36 μs after discharge initiation. To capture
this enhanced fluorescence, the sensitivity of the detection electronics was de-
creased by a factor of 45. When the Mn plasma was created without a C discharge,
Fig. 3(d) shows that there is indeed some spurious radiation at 2177 Å (line or con-
tinuum) from the Mn plasma which is detected. When this spurious background is sub-
tracted from the fluorescence in Fig. 3(c), the net enhanced fluorescence is 180
times the spontaneous emission, i.e., optical pumping increases the 4p $^1P^0$ popula-
tion by a factor of 180. Such enhancements were found to be very reproducible over
hundreds of shots. The typical duration of the enhanced fluorescence (FWHM) was
about 0.5-1 μs.

Similar enhancements were then made at several other n = 4-3 wavelengths in
CIII. The net enhanced fluorescence on the 4s ^1S - 3p $^1P^0$ line at 1894.5 Å, and the

4f $^1F^0$ - 3d ^1D line at 2163 Å was found to be about a factor of 70 above the sponta-
neous emission. Enhanced fluorescence was also measured on the 4d ^3D - 3p $^3P^0$ line
at 1620 Å and the 4f $^3F^0$ - 3d ^1D line at 1923 Å. These measurements confirm that
the photoexcitation of the 4p $^1P^0$ state is accompanied by rapid collisional exchange
of this population with other n = 4 levels.

The CII, 4p $^2P^0$ - 3s ^2S line at 2174 Å and the CIII, 2p^2 $^1P^0$ - 2s2p ^1S line at
2297 Å were also studied. No fluorescence was observed, confirming the selectivity
of the resonant photoexcitation. As an additional check, a laser-produced Al plasma
was substituted for the Mn plasma. No fluorescence was observed at any of the CIII,
4-3 wavelengths, thus ruling out collisional excitation by electrons from the laser-
produced plasma, or broadband photoexcitation. These fluorescence measurements con-
firmed the general predictions [10] of the kinetics of resonant photoexcitation in
CIII. To further quantify the kinetics and to estimate small-signal gain under
optimum CIII discharge conditions, a detailed collisional-radiative model was
developed. This model is described in the next section.

III. COLLISIONAL-RADIATIVE MODEL FOR CIII

A collisional-radiative model for CIII was developed in two steps. First, the
relative abundances of different ground state populations were calculated, ignoring
all excited states. Then, using the calculated ground states of CIII and CIV, a 72-
level model was constructed for CIII. Such an approach is valid when the excited
state populations are very small relative to the ground states. Electron density n_e
and T_e are essential inputs to such a model. n_e and T_e were measured spectroscopi-
cally at five axial locations up to 25 mm from the cathode. Density was in the
range $6 \times 10^{15} \leq n_e \leq 1.2 \times 10^{16}$ cm^{-3}. T_e was uniform at 4 eV. These measurements
will be described elsewhere. Processes considered in the ionization balance includ-
ed collisional ionization [11], three-body recombination [12], radiative recombina-
tion [13], and dielectronic recombination [14,15]. For $n_e = 10^{16}$ cm^{-3} and $T_e =$
4 eV, 70% of the ions are in the CIII ground state, and 30% in the CII ground state.

Starting with the ground state populations of CIII and CIV, a 72-level, colli-
sional-radiative model was developed for CIII. In this model, all singlet and trip-
let states of the outer electron configurations, with principal quantum numbers
n = 2 to n = 6 were included. For n = 7 to 10, single lumped states distinguished
only by principal quantum number were included. For all levels considered from
n = 2 to 6, energy levels, transition oscillator strengths and collisional excita-
tion/de-excitation rates were provided by Dr. W.L. Morgan of Lawrence Livermore Lab-
oratory. For n = 7 to 10 levels, hydrogenic rates [16] were used. The generalized
rate equation for the jth level is as follows.

$$\frac{dn(j)}{dt} = -n(j)[S(j)n_e + \sum_{j>i} A(j,i) + \sum_{j>i} C^d(j,i)n_e + \sum_{j<i} C^e(j,i)n_e]$$
$$+ n(i)[\sum_{j<i} A(i,j) + \sum_{j<i} C^d(i,j)n_e + \sum_{j>i} C^e(i,j)n_e]$$
$$+ n_g^{CIV}[\alpha(j) + \beta(j)n_e] \tag{1}$$

n_g^{CIV} is the CIV ground state density, S(j) is the collisional ionization rate
[17-19], $C^d(j,i)$ and $C^e(j,i)$ are the collisional de-excitation and excitation rates,
$\beta(j)$ is the radiative recombination rate [20], and $\alpha(j)$ is the three-body recombina-
tion rate, which was determined from the ionization rate by detailed balancing. For
$n_e > 10^{15}$ cm^{-3}, some of the lower lying (resonance) lines can be optically trapped.
Holstein escape factors [21,22] were used to account for the effects of opacity on
all levels. Since the relaxation times for excited levels are much shorter than the
time scales for evolution of the CIII and CIV ground states, the excited state popu-
lations attain quasi-steady-state values.

Figure 4(a) shows typical quasi-steady-state level populations vs n_e. The
dashed lines on the figure show the absolute population density of the 4s, 4p, and
4f singlet states as a function of n_e, for a fixed $T_e = 4$ eV. The full curves on
the figures are the enhanced populations of these levels, when the 4p $^1P^0$ level is
resonantly photoexcited by MnVI line radiation. The absolute intensity of the MnVI
pump line was not measured. Based on measurements of absolute continuum intensity
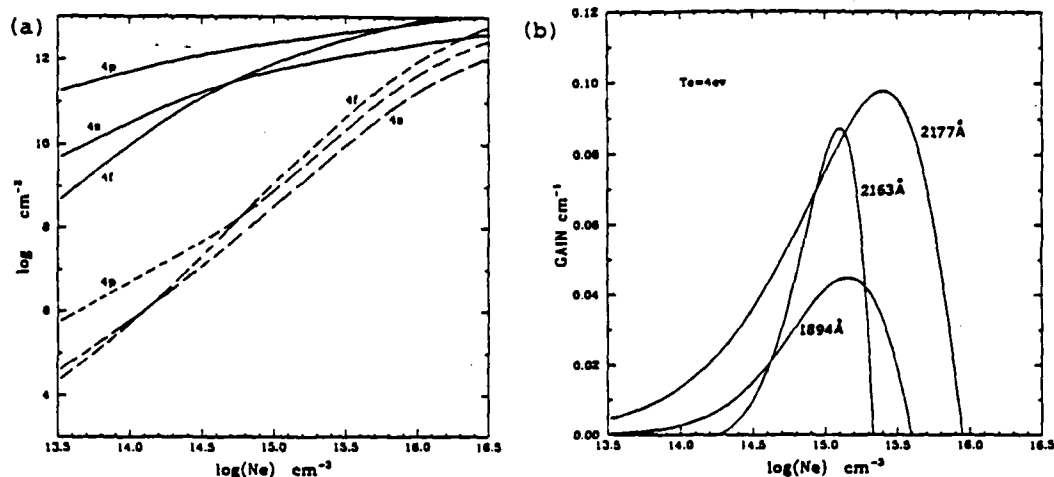
Fig. 4. Theoretical calculations. (a) Populations of the 4s, 4p, and 4f singlet
levels in CIII vs n_e for $T_e = 4$ eV (dashed curves); enhanced populations of
4s, 4p, and 4f when the 4p $^1P^0$ level is photoexcited (full curves). The
MnVI pump line was assumed to have a brightness temperature of 20 eV.
(b) Predicted small-signal gain coefficients vs n_e for the 2177, 2163, and
1894 Å lines in CIII ($T_e = 4$ eV).

and a self-similar hydrodynamic expansion model, the initial Mn plasma temperature
was estimated to be 35 eV. The brightness temperature of the MnVI pump line will be
somewhat lower, and was assumed to be 20 eV. The effective solid angle for photoex-
citation was assumed to be π steradians. With this brightness temperature and as-
suming Doppler line profiles for MnVI and CIII, stimulated absorption and emission
were included in Eq. 1 for the 4p level, to give the enhanced n = 4 populations in
CIII. As shown by the full curves in Fig. 4(a), significant enhancement occurs for
all $n_e < 10^{16}$ cm^{-3}. It should be recalled that the measured value of n_e in the CIII
discharge at a current of 6400 A was about 7×10^{15} cm^{-3} over the first 25 mm from
the cathode. For this value of n_e, Fig. 4 shows a population enhancement of 4p by
about a factor of 140 while the 4f and 4s levels are enhanced by about a factor of
70 and 90, respectively, over their spontaneous values. These predicted enhance-
ments agree remarkably well with the measurements described earlier, over the same
distance downstream from the cathode.

For n_e between 10^{14} cm^{-3} and 10^{16} cm^{-3}, the enhanced n = 4 populations are high
enough to invert them with respect to the n = 3 levels. Gain is thus possible on
many 4-3 transitions. Typical small-signal gains are plotted in Fig. 4(b), vs n_e,
for a fixed T_e of 4 eV. The gains shown are for the 4s-3p, 1894.5 Å, 4p-3d, 2177 Å,
and 4f-3d, 2163 Å lines. At the densities actually observed in discharges of about
6400 A, gains of about 0.1 cm^{-1} are predicted for 2163 and 2177 Å. If the 23 mm
long Mn plasma provided uniform photoexcitation over a 20 mm length of the CIII dis-
charge, overall, single-pass gains of 0.2, or 22%, were expected. Single-pass gains
of 100% per pass, with a gain coefficient of 0.4 cm^{-1}, were measured at both the
2177 and 2163 Å wavelengths, as described in the next section.

IV. SINGLE-PASS GAIN MEASUREMENTS

For gain measurements, the mirror behind the hollow anode (see Fig. 2) was
adjusted to reflect rays from the pumped region of the CIII plasma back onto them-
selves. Irises were placed on either side of the discharge, to ensure that the
solid angles subtended by the reflecting mirror and the spectrometer gathering
optics were identical. On successive shots, the spontaneous emission and enhanced
fluorescence were measured with and without the reflecting mirror. The ratio R of
the measured line intensities with and without the mirror is given by:

$$R = 1 + re^{-\tau} \tag{2}$$

where r is the net reflectivity of the vacuum window and reflection mirror, and τ
is the integrated absorption coefficient along the entire optical path length. For

CIII 4p'P°– 3d'D 2177Å

CIII 4f'F°– 3d'D 2163Å

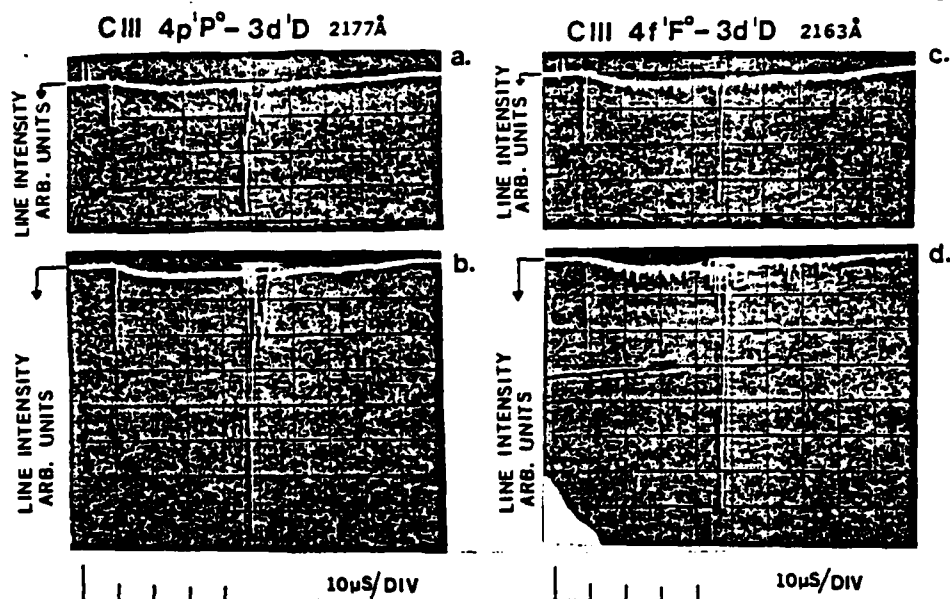10μS/DIV   10μS/DIV

Fig. 5. Single-pass gain at 2177 and 2163 Å in CIII. (a) Enhanced fluorescence
at 2177 Å, without reflecting mirror; (b) fluorescence at 2177 Å, with
reflecting mirror; (c) enhanced fluorescence at 2163 Å, without reflecting
mirror, (d) fluorescence at 2163 Å, with reflecting mirror.

non-inverted lines, $\tau \geq 0$, hence $R \leq (1 + r)$. For an inverted transition with gain,
$\tau < 0$ and $R > (1 + r)$. To calibrate the single-pass reflection optics, the CII line
at 2174 Å was first examined. The measured value of R was 1.5, which implies $r =$
0.5 from Eq. 2 (since $\tau = 0$ for this line). In addition, the spontaneous emission
(without photoexcitation) at 2163 A in CIII was similarly measured. Again R = 1.5,
or $r = 0.5$. R was then measured at 2177 Å and 2163 Å with photoexcitation, to
measure the gain τ.

Figure 5 shows the measurement of R with the fluorescence at 2177 Å and 2163 Å,
and evidence for gain. Figure 5(a) shows the enhanced fluorescence at 2177 Å, with-
out the reflecting mirror. The spontaneous emission is a factor of 170 smaller than
this enhanced fluorescence, and so is buried in the baseline. The slight baseline
shift is due to a ground-loop in the discharge circuit. Figure 5(b) shows the fluo-
rescence with the reflecting mirror. The ratio R is 2.1. For $r = 0.5$, this gives
$\tau = 0.8$. For a 2 cm effective gain length in the CIII plasma, this gives a gain
coefficient of 0.4 cm^{-1}. Figures 5(c) and 5(d) show similar results at the CIII,
2163 Å wavelength. For this line, R = 2.1 also, and hence the gain coefficient is
also 0.4 cm^{-1}.

The measured gain coefficients are significantly higher than those predicted by
the collisional-radiative model (see Fig. 4). The discrepancy could be due to over-
estimation of the effects of opacity in the model. Streaming motion of the ions,
smaller lateral plasma dimension than that assumed, and the effect of gradients will
all reduce the opacity effects and correspondingly increase the predicted gains.

V. SUMMARY

A detailed theoretical and experimental study was presented of resonant photo-
excitation in CIII ions by MnVI line radiation. Using an optimized discharge and
pump geometry, enhanced fluorescence and small-signal gain on the 4p-3d, 2177 Å and
the 4f-3d, 2163 Å transitions in CIII were measured. Gain coefficients of 0.4 cm^{-1}
were measured for both wavelengths. To compare with the experiments, a detailed,
72-level, collisional-radiative model was developed for CIII. An ionization balance
first gave the CIII and CIV ground state populations, using measured values of n_e
and T_e. These ground state populations in turn yielded the populations of various
excited states in CIII. Resonant photoexcitation was modeled by an assumed bright-
ness temperature of 20 eV for the pump line. Good agreement was obtained between

experiment and theory. The high gains measured and their ~1 μs duration suggest that a Fabry-Perot cavity may be used to demonstrate laser oscillation at the 2177 and 2163 Å wavelengths. Such experiments are under way and will be reported elsewhere, along with further details on the experimental measurements and the atomic physics code.

VI. ACKNOWLEDGMENTS

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LASER OSCILLATION AT 2177 AND 2163Å IN CIII,

RESONANTLY PHOTO-EXCITED BY 310Å, MnVI LINE RADIATION

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PACS numbers: 42.55.Hq, 32.30.Jc, 32.50.+d, 32.80.Bx

ABSTRACT

Laser oscillation is reported at 2177 and 2163Å in CIII ions, in a vacuum-arc discharge, pumped by MnVI line radiation from a laser produced Mn plasma. This Be-like UV laser pumped by resonant photoexcitation is a prototype for soft x-ray lasers in higher Z, isoelectronic analogs.

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Resonant photoexcitation of one ion by intense line radiation from another ion, is one method of creating a population inversion. Efficient pumping requires that the wavelength mismatch between the pump and pumped transitions be small compared to their linewidths. As early as 1930, enhanced fluorescence at 3.2μ in CsI was observed[1], using 3880Å pump line radiation from HeI. In 1961, such resonant photoexcitation led to laser oscillation[2] at 3.2μ. The concept was extended[3] to 5461Å in HgI, using two-step photoexcitation.

Many soft x-ray laser schemes were proposed[4-7] using resonant photoexcitation, in H-like and He-like ions. Recently a new class of photoexcited lasers was identified[8] in Be-like ions, with laser wavelengths from 2177Å in CIII pumped by MnVI, to 200Å in MgIX pumped by AlXI. The basic concept is shown in Fig. 1. MnVI line radiation at 310.182Å pumps the $2s^2$ ^1S - $2s4p$ $^1P^0$ transition at 310.170Å in CIII. The 4p population is inverted with respect to the 3d level and a laser is possible on the 4p $^1P^0$-3d ^1D transition at 2177Å. This scheme was studied experimentally. CIII ions in a vacuum-arc discharge were photoexcited by MnVI line radiation from an adjacent, laser produced plasma. Electron collisions within the CIII discharge transferred some of the excited 4p population to other n=4 levels. Enhanced fluorescence was measured[9] on several 4-3 transitions at 1620, 1894.5, 1923, 2163 and 2177Å. This Letter reports measurements of single-pass gain and laser oscillation at 2177 and 2163Å in CIII, pumped by

MnVI line radiation. This is believed to be the first UV laser pumped by resonant photoexcitation. By isoelectronic scaling of this prototype laser, shorter wavelength lasers can be produced in higher Z analogs. Soft x-ray amplifiers have recently been demonstrated[10,11] in laser produced plasmas pumped by electron collisions and by recombination.

Figure 2 is a schematic drawing of the experimental apparatus. The CIII ions are produced in a vacuum-arc discharge between a 6mm diameter carbon cathode and a hollow carbon anode 100mm apart. A discharge is triggered by focusing a 15J/100ns CO_2 laser on to the cathode face. The discharge current has a 6.4kA peak and 60μs flat-topped duration as shown in figure 3a. The MnVI pump plasma is produced by focusing another 15J/100ns CO_2 laser to a 2 mm x 23 mm line focus on a Mn slab, as shown.

In the first experiments, single-pass gain in CIII was measured at 2177 and 2163Å. The front mirror of the laser cavity (labelled A in Fig. 2) was replaced by a lens (shown inset in Fig.2) which collimated and focused the pumped CIII region on to the entrance slit of the UV monochromator. On successive shots, the spontaneous emission and enhanced fluorescence were measured with and without the reflecting mirror B. The ratio R of the measured line intensities with and without the mirror is given by:

$$R = 1 + se^{-\tau} \qquad\qquad (1)$$

where τ is the integrated absorption coefficient along the entire optical path length, and $s(=0.5)$ is the measured net reflectivity of the vacuum window and mirror B. For an inverted transition with gain, $\tau<0$ and $R>(1+s)$. At both 2177 and 2163$\mathring{A}$, the measured value of R (for 1μs coincident with the Mn plasma) was 2.1. For $s=0.5$, this gives $\tau=-0.8$. For a 2cm gain length, this gives a gain coefficient of 0.4cm^{-1}. To check this, the CII line at 2174$\mathring{A}$ was then examined. This CII line was measured to be optically thin ($\tau=0$) and occurs conveniently close to the two lasing wavelengths. The measured value of R for CII was 1.5, thus verifying the gain on the CIII lines. This small signal gain is similar to the prediction of a 72-level collisional-radiative model[12] for photoexcitation. With measured values of electron density and temperature in the C discharge, populations of n=3 and 4 levels were calculated using atomic data provided by W.Morgan[13]. Photoexcitation was included by estimating[12] a 20eV brightness temperature for the MnVI pump line. The predicted single-pass gain was 0.15cm^{-1} at both 2177 and 2163$\mathring{A}$.

Laser oscillation at 2177$\mathring{A}$ and 2163$\mathring{A}$ was verified as follows: The collimating lens (C in Fig.2) was replaced by the front mirror A of a confocal, Fabry-Perot cavity. The mirrors A and B had 1m radius, were spaced 2m apart, and had reflectivity r=0.94 at the laser wavelengths, while the two Brewster windows

had transmission t=0.96. First,the UV monochromator was tuned to a wing(optically thin) of the CIII, $2p^2$-2s2p line at 2297Å. The upper level $2p^2$ of this line has no allowed transitions to either the upper or lower laser levels. No fluorescence there-fore occurs on this line, which serves to check the evidence for lasing. Figure 3b shows the measured cavity output of the spontaneous emission from this line, at a discharge current of 6.4kA. The Mn pump plasma was produced 36μs after carbon dis-charge initiation as indicated by the "noise" fiducial on all three traces in Fig.3. No enhanced fluorescence is observed at this wavelength, as expected. Figure 3c shows the spontaneous emission, when mirror A was removed. The measured signal is larger, since the partially transmitting mirror A was removed from the line of sight. For a perfectly aligned cavity, it is straightforward to derive an expression for the ratio of the measured line intensity with the mirror A removed, I_1, to that with the tuned cavity, I_2, viz:

$$\frac{I_1}{I_2} = \frac{1-r^2 t^4 e^{2\alpha}}{(1-r)\{1-(rt^2 e^{\alpha})^{N+1}\}} \qquad (2)$$

where α is the gain coefficient, N the number of passes, and r and t were defined earlier. At 2297Å, α=0, and Eq. (2) gives I_1/I_2 =4.1, for large N (>200) which compares well with the measured ratio from Fig. 3 of about 4. When α>0, lasing requires $rt^2 e^{\alpha} \gtrsim 1$. Then Eq. (2) shows that I_1/I_2<1, for large N i.e. re-

moving the mirror A will actually cause a decrease in the detect-
ed signal, rather than an increase. Such a decrease was observed
at 2177 and 2163Å. Figure 4a shows the measured emission at
2177Å, with the tuned laser cavity. On these scales, no spontane-
ous emission is observed before and after photoexcitation, and
strong emission occurs coincident with the MnVI pump radiation.
When mirror A was removed, Fig. 4b shows that the detected emis-
sion decreased, to about 30% of the value with the cavity. This
decrease confirms laser oscillation at 2177Å. The spontaneous
emission level before and after optical pumping in Fig. 4a is up
to a factor of 1000 below the laser intensity. Similar results
were obtained at 2163Å, on the $4f\ ^1F^0 - 3d\ ^1D$ line. Figure 4c
shows the laser intensity with the tuned cavity. Figure 4d shows
a decrease in intensity, to 36%, when mirror A was removed.
Again, this decrease confirms laser oscillation at 2163Å. As a
further check on these measurements, the laser beam which pro-
duced the Mn plasma was partially blocked to reduce the gain
length to about 1cm. The ratio I_1/I_2 increased to 0.8 at 2177Å
and 0.9 at 2163Å. This increase in the ratio corresponds to
lower gain α, but still indicates lasing. Wide bandwith elec-
tronics showed that the laser duration was about 0.5-1μs, corre-
sponding to between 75 and 150 passes in a stable cavity.

Suppose that the saturated gain α at the two laser wave-
lengths, is such that $rt^2e^{\alpha} = 1 + \epsilon$, where $\epsilon \ll 1$ i.e. the laser
is just above threshold. Then Eq. (2) reduces to :

$$\frac{I_1}{I_2} \simeq \frac{2}{(1-r)(N+1)} \tag{3}$$

For N=100, I_1/I_2=0.33. This predicted value compares very well with the measured values of 0.3 and 0.36 at 2177Å and 2163Å, respectively.

A rough estimate of the power and energy/pulse from these photoexcited lasers is obtained by assuming that the saturated gain coefficient is about $0.1cm^{-1}$ (the measured small signal gain was ~ $0.4cm^{-1}$). This gives an upper level population (4f) of ~ $10^{12}cm^{-3}$. Within an active volume 1.0cm in diameter x 2.5cm long, the estimated laser power output at 2163Å is 1.5kW. For a 1μs laser, the energy per pulse is 1.5mJ. For a 6.4kA discharge current, with a 50mΩ plasma impedance the electrical energy input over 1μs is 2J. The laser energy input to the Mn plasma is ~ 10J. The laser is not strictly necessary, as a fast capacitive discharge with ~ 10J could also be used to produce the Mn pump radiation, with high efficiency. The overall efficiency of these photoexcited lasers is thus ~ 1.5mJ/12J ~ 10^{-4}. It is possible that soft x-ray analogs of the CIII-MnVI laser described here could be produced, with similar overall efficiencies. It should be pointed out that the experimental configuration used is far from optimal. By making the pump plasma coaxial with the laser medium, a greater solid angle can be utilized for photoexcitation, increasing the gain, and hence the efficiency.

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FIGURE CAPTIONS

Fig.1 Energy levels in CIII. Optical pumping of the $2s^2\ {}^1S$ - $2s4p\,{}^1P^0$ transition is accompanied by collisional transfer to the 4s, 4d, and 4f levels. Lasing is possible on the 4s-3p, 4p-3d, and 4f-3d transitions.

Fig.2 Schematic diagram of the experimental apparatus.

Fig.3 Discharge current and CIII 2297Å spontaneous emission vs. time. 3(a) current 2.3kA/div. (noise pickup marks the firing of the CO_2 laser to produce the Mn plasma, 36µs after initiation of the CIII discharge). 3(b) CIII 2297Å line emission, with a tuned cavity. 3(c) CIII 2297Å line emission with the front cavity mirror A removed.

Fig.4 Evidence for lasing in CIII. 4(a) Emission at 2177Å, with laser cavity. 4(b) Emission at 2177Å with the front cavity mirror removed. 4(c) Emission at 2163Å, with the laser cavity. 4(d) Emission at 2163Å, with the front cavity mirror removed.
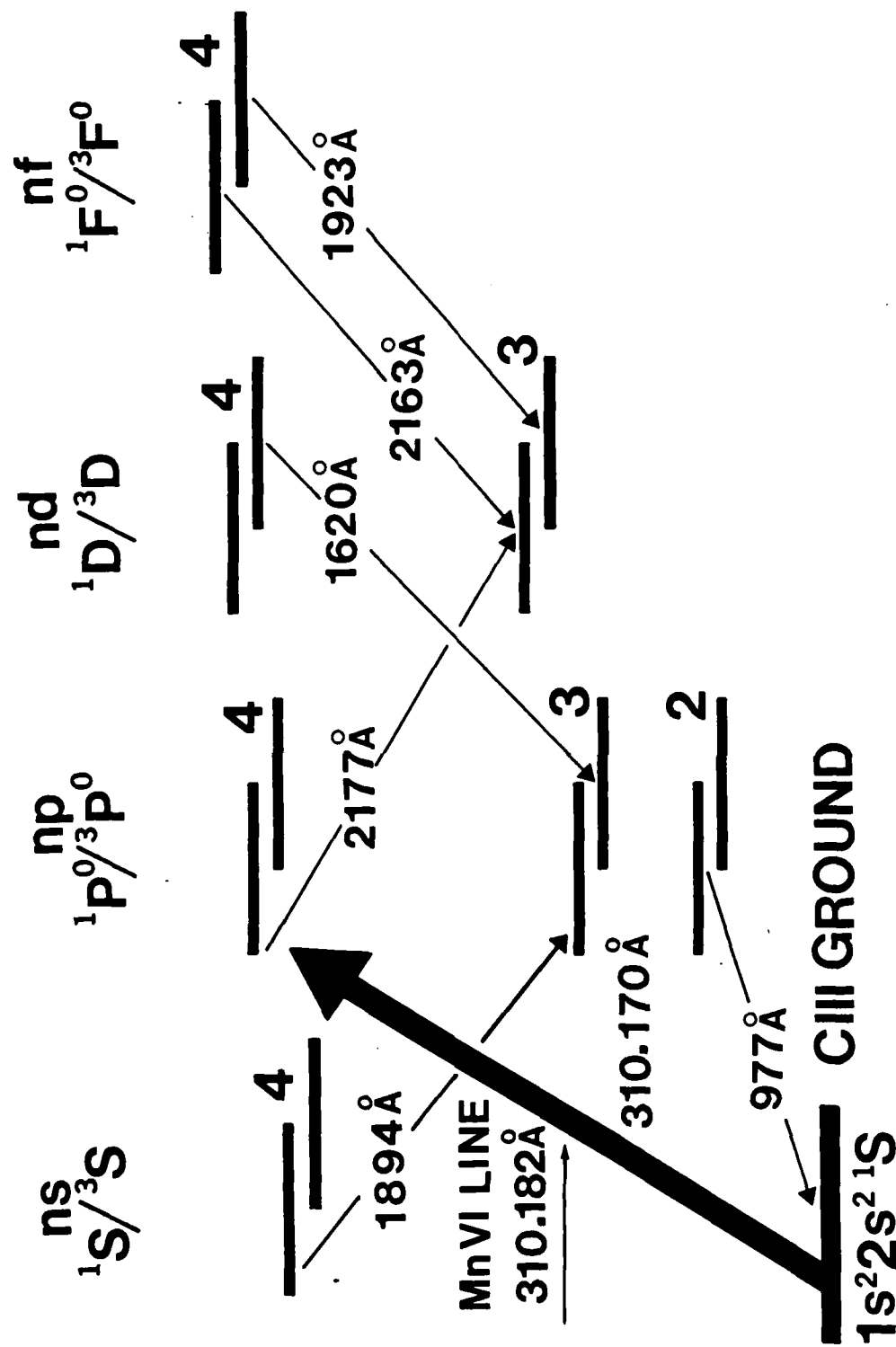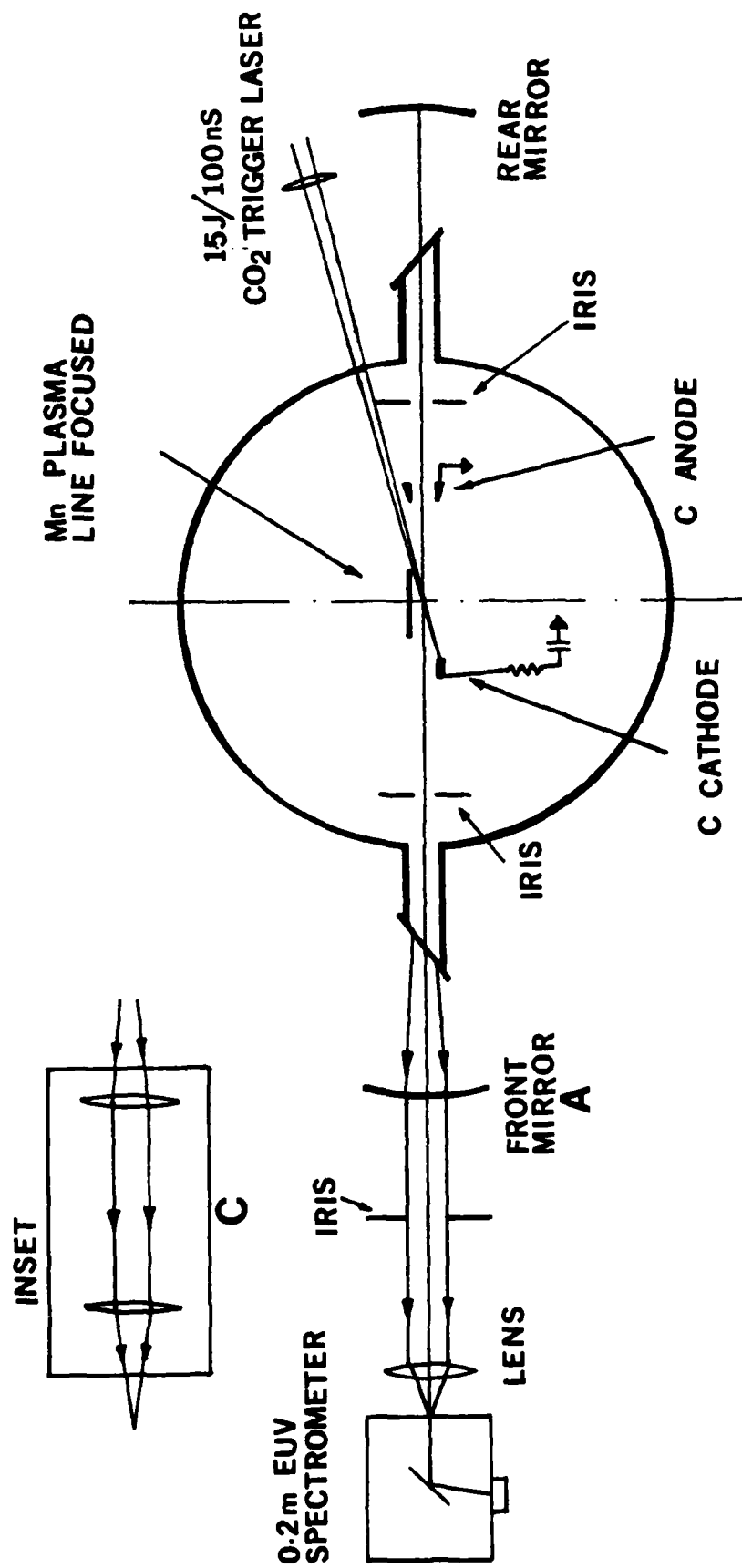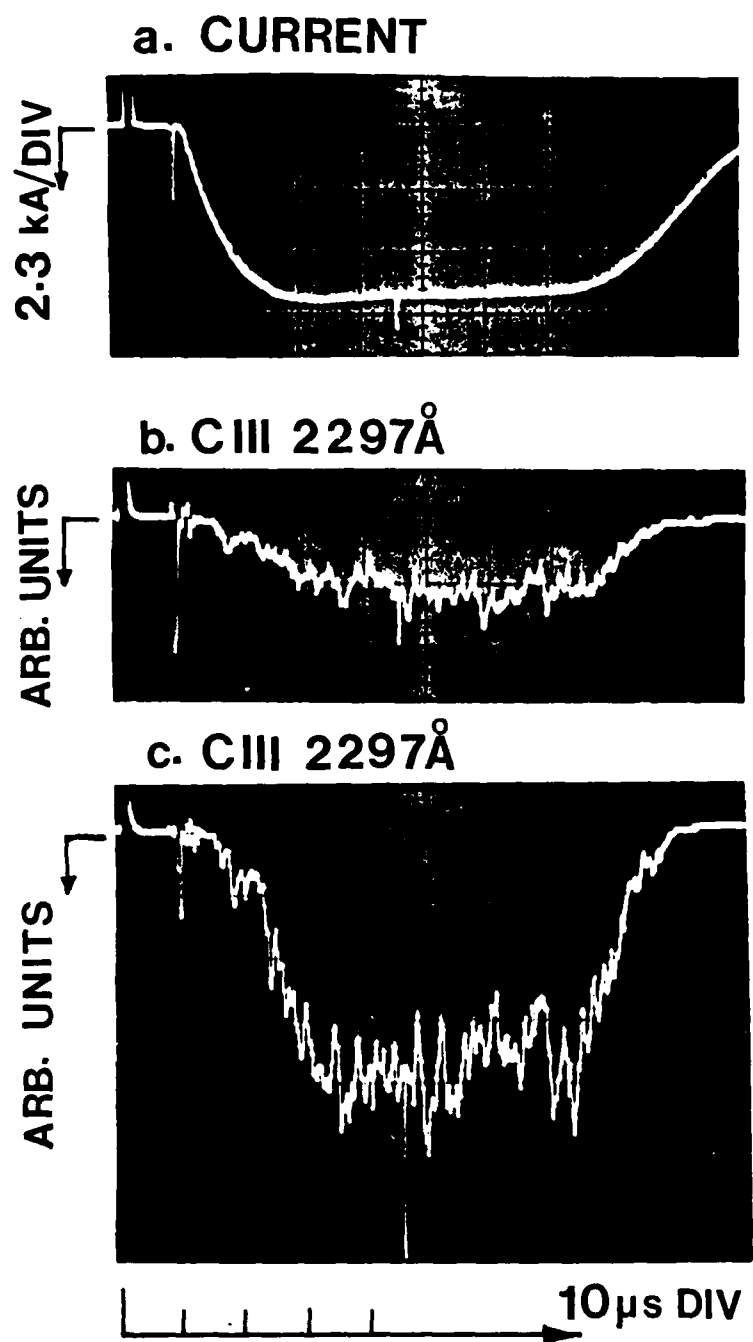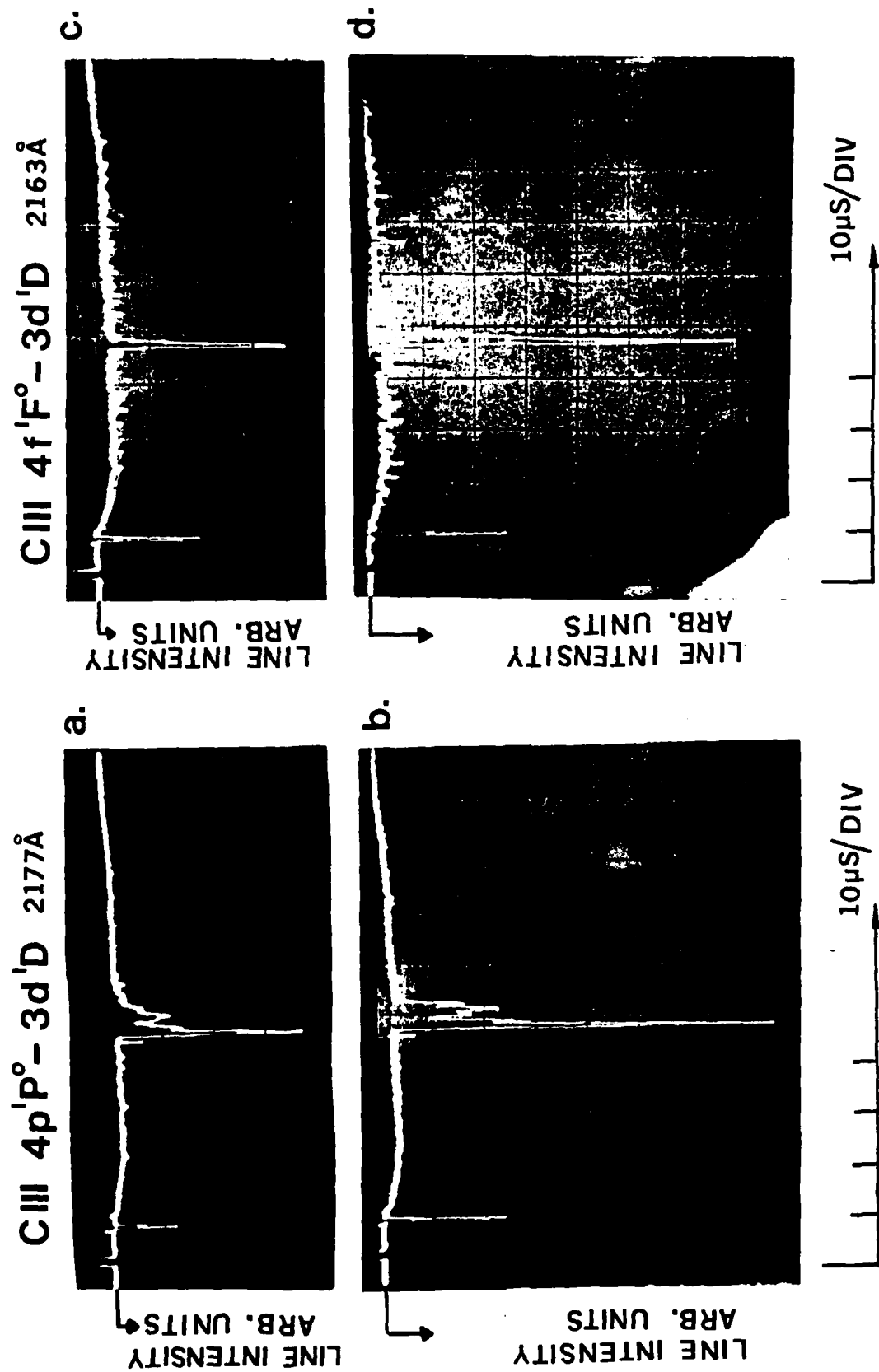
Fig. 1

FIG. 2

a. CURRENT

2.3 kA/DIV

b. CIII 2297Å

ARB. UNITS

c. CIII 2297Å

ARB. UNITS

10 μs DIV

Fig. 3

CIII 4f'F° – 3d'D 2163Å

CIII 4p'P° – 3d'D 2177Å

10μS/DIV

10μS/DIV

LINE INTENSITY ARB. UNITS

Fig. 4

END
DTIC

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